

Influence of Cellulose Nanocrystals and Surfactants on Catastrophic Phase Inversion and Stability of Emulsions

by

Hyungseak Kim

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Author's Declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

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Abstract

Emulsions are thermodynamically unstable colloidal systems that play a critical role in numerous industrial applications, including food, cosmetics, pharmaceuticals, and enhanced oil recovery. A key limitation of emulsions is their susceptibility to destabilization phenomena such as coalescence, creaming, sedimentation, and Ostwald ripening. Phase inversion, the transition of a water-in-oil (W/O) emulsion to an oil-in-water (O/W) system, or vice versa, represents a particularly important instability mechanism. The two primary mechanisms of emulsion phase inversion are transitional and catastrophic. Catastrophic phase inversion is driven by the increasing volume fraction of the dispersed phase and often results in abrupt and irreversible structural changes that compromise emulsion stability. Although the phase inversion of surfactant-stabilized emulsions has been extensively studied, comparatively little is known about how particle stabilizers such as nanocrystalline cellulose (NCC) influence catastrophic inversion behavior.

This thesis presents a systematic investigation of the catastrophic phase inversion and coalescence stability of emulsions stabilized by NCC and compares these systems directly with emulsions stabilized by molecular surfactants of different headgroup chemistries: anionic (sodium dodecyl sulfate, SDS), cationic (octadecyltrimethylammonium chloride, OTAC), nonionic (C12-14 alcohol ethoxylate, Alfonic®), and zwitterionic (cetyl betaine, Amphosol®). Phase inversion was monitored through electrical conductivity measurements under controlled emulsification conditions, while coalescence stability and interfacial properties were evaluated using storage experiments and pendant drop tensiometry, respectively.

The results demonstrate that NCC markedly delays catastrophic phase inversion by increasing the critical aqueous phase volume fraction required for inversion, rising from 0.253 in the absence of NCC to 0.545 at 1.5 wt% NCC. This behavior is attributed to interfacial jamming and the formation of rigid particle-laden films that suppress droplet coalescence and deformation. In contrast, surfactant-stabilized systems exhibit a non-monotonic inversion response: low concentrations delay inversion through limited interfacial coverage, whereas higher concentrations promote earlier inversion due to interfacial saturation and curvature stabilization of O/W emulsions. Surface and interfacial tension measurements confirmed the weak surface activity of NCC relative to all

surfactants tested, yet NCC imparted superior coalescence resistance at concentrations above 0.2 wt%.

The findings of this work highlight the distinct mechanisms by which surfactants and particles govern catastrophic phase inversion. While surfactants reduce interfacial tension and dynamically redistribute at interfaces, NCC stabilizes emulsions through irreversible adsorption, steric hindrance, and network formation. To the best of our knowledge, this thesis provides the first quantitative comparison of NCC- and surfactant-stabilized catastrophic inversion behavior under identical conditions. The outcomes contribute to new mechanistic insights into the stabilization of high internal phase emulsions and highlight the potential of NCC as a sustainable, bio-based alternative to synthetic surfactants in industrial formulations.

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I would like to express my sincere gratitude to my supervisor, **Professor Rajinder Pal**, for his guidance, encouragement, and support throughout the course of this research. His expertise and insight were invaluable in shaping this work and in my development as a researcher.

I would also like to acknowledge **Walker Industries** for their support, particularly for providing the flexibility and time away from work that enabled me to pursue graduate studies. Their encouragement made this academic journey possible.

Dedication

I am also deeply thankful for the unwavering support of my family. To my wife, **Alaya**, for her love, patience, and encouragement; to my son, **Jio**, whose joy and curiosity inspire me daily; and to my son, **Jin**, whose arrival has been a constant source of motivation. Their support has been the foundation that enabled me to complete this thesis.

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List of Abbreviations

Abbreviation	Full Form / Description
AFM	Atomic Force Microscopy
CMC	Critical Micelle Concentration
CNF	Cellulose Nanofibril
CNC	Cellulose Nanocrystal
CPI	Catastrophic Phase Inversion
DLVO	Derjaguin–Landau–Verwey–Overbeek theory
EIP	Emulsion Inversion Point
EO	Ethylene Oxide
EOR	Enhanced Oil Recovery
HLD	Hydrophilic–Lipophilic Difference
HLD–NAC	Hydrophilic–Lipophilic Deviation–Net Average Curvature
HLB	Hydrophilic–Lipophilic Balance
IFT	Interfacial Tension
kT	Thermal Energy Unit (Boltzmann constant $\times$ temperature)
NCC	Nanocrystalline Cellulose
O/W	Oil-in-Water
OTAC	Octadecyltrimethylammonium Chloride
PIT	Phase Inversion Temperature
SEM	Scanning Electron Microscopy
SFT	Surface Tension
SDS	Sodium Dodecyl Sulfate
TEM	Transmission Electron Microscopy
TPI	Transitional Phase Inversion

Abbreviation	Full Form / Description
W/O	Water-in-Oil

List of symbols

Symbol	Description / Definition	Units
A	Interfacial area created during emulsification	m ²
α	Temperature coefficient in the HLD equation	—
a_0	Correlation length (interface smoothness scale)	nm
C	Surfactant concentration	mol·L ⁻¹
C*	Critical micelle concentration (CMC)	mol·L ⁻¹
ΔG	Gibbs free energy of emulsification	J·mol ⁻¹
γ	Interfacial tension (oil–water)	mN·m ⁻¹
Γ_i	Surface excess concentration of component <i>i</i>	mol·m ⁻²
φ	Volume fraction of dispersed phase	—
k	Boltzmann constant	J·K ⁻¹
kT	Thermal energy (Boltzmann constant × temperature)	J
L_s	Characteristic surfactant length	nm
μ_i	Chemical potential of component <i>i</i>	J·mol ⁻¹
n	Number of ethylene oxide (EO) units	—
ρ	Density of the continuous or dispersed phase	kg·m ⁻³
R	Droplet radius	μm
σ	Interfacial tension (alternative notation for γ)	mN·m ⁻¹

Symbol	Description / Definition	Units
t	Time (used in coalescence and stability studies)	s
T	Absolute temperature	K
τ	Shear stress / Emulsification torque	Pa
θ	Contact angle at oil–water–solid interface	° (degrees)
V	Volume of dispersed or continuous phase	m ³
η	Viscosity of continuous phase	Pa·s
ψ	Surface potential	mV
χ	Interaction parameter (HLD contribution from oil type)	—
λ	Interfacial rigidity or bending modulus	N·m
HLD	Hydrophilic–Lipophilic Difference value	—
HLD₀	Balanced HLD condition (HLD = 0)	—

Chapter 1. Introduction

1.1. Background on Emulsions and Colloidal Systems

Emulsions are among the most versatile and widely studied colloidal systems, underpinning technologies that span from everyday consumer products to advanced industrial processes. By definition, an emulsion is a heterogeneous dispersion of two immiscible liquids, in which one phase is dispersed as fine droplets within the other. Despite their apparent simplicity, emulsions exhibit complex physicochemical behavior that arises from the large interfacial area separating the two phases and the intricate balance of forces that determine droplet stability [1,2]. This complexity makes emulsions both a focus of fundamental scientific study and a foundation for a wide range of technological applications. They are central to the formulation of foods, cosmetics, pharmaceuticals, coatings, and enhanced oil recovery fluids, yet their usefulness is continually challenged by inherent instability and eventual destabilization through coalescence, creaming, or phase inversion. A deeper understanding of the fundamental science of emulsions is therefore crucial for improving stability, tailoring properties, and expanding their application across diverse fields of chemical engineering and materials science.

Colloidal systems such as emulsions are encountered in both daily life and industrial practice. In the food sector, examples include milk, mayonnaise, and salad dressings; in personal care, lotions, creams, and shampoos; in pharmaceuticals, emulsions are employed for drug delivery and controlled release; and in the energy sector, petroleum-based emulsions play a role in crude oil transport and enhanced oil recovery [3,4]. Their significance is also evident in construction materials, where asphalt emulsions are widely used in road surfacing and concrete admixtures to improve durability and reduce processing energy [5,6], and wax emulsions are incorporated into paper packaging and wood-based composites to provide water resistance and barrier properties [7]. Emulsions also function as essential intermediates in processes such as polymerization, coatings, and encapsulation, reinforcing their role as a versatile platform technology across the chemical and materials industries. [1]

The stability of emulsions is dictated by interfacial phenomena. The high interfacial energy between immiscible liquids creates a strong thermodynamic drive for droplets to coalesce and reduce interfacial area and energy. Stabilizers, typically surfactants or colloidal particles, adsorb

at the oil–water interface to counteract this tendency by lowering interfacial tension, imparting steric or electrostatic barriers to droplet coalescence, or forming mechanically rigid interfacial layers [1,8,9]. The selection of stabilizer type, together with droplet size distribution and processing conditions, ultimately governs emulsion morphology, lifetime, and performance.

Emulsions are commonly classified according to the identity of the continuous phase. In oil-in-water (O/W) emulsions, oil droplets are dispersed in an aqueous medium, whereas in water-in-oil (W/O) emulsions, water droplets are dispersed in oil. More complex structures, such as double emulsions (W/O/W and O/W/O) and bicontinuous emulsions, further expand the diversity of emulsion morphologies and functions [10]. Schematics of simple, multiple, and bicontinuous emulsions are shown in Figures 1 and 2 [11,12]. Emulsion type is governed by formulation parameters, including the chemistry of the stabilizer, the relative volume of the phases, the intensity of mixing, and the curvature of the interface.

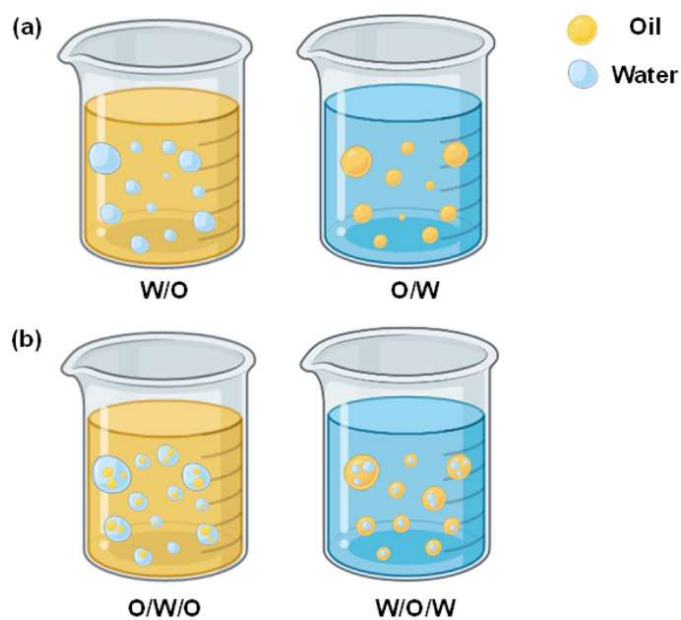


Figure 1. Schematic of a) simple emulsion types (W/O) and (O/W) systems, and b) multiple emulsions (O/W/O) and (W/O/W) systems reproduced from reference [12]

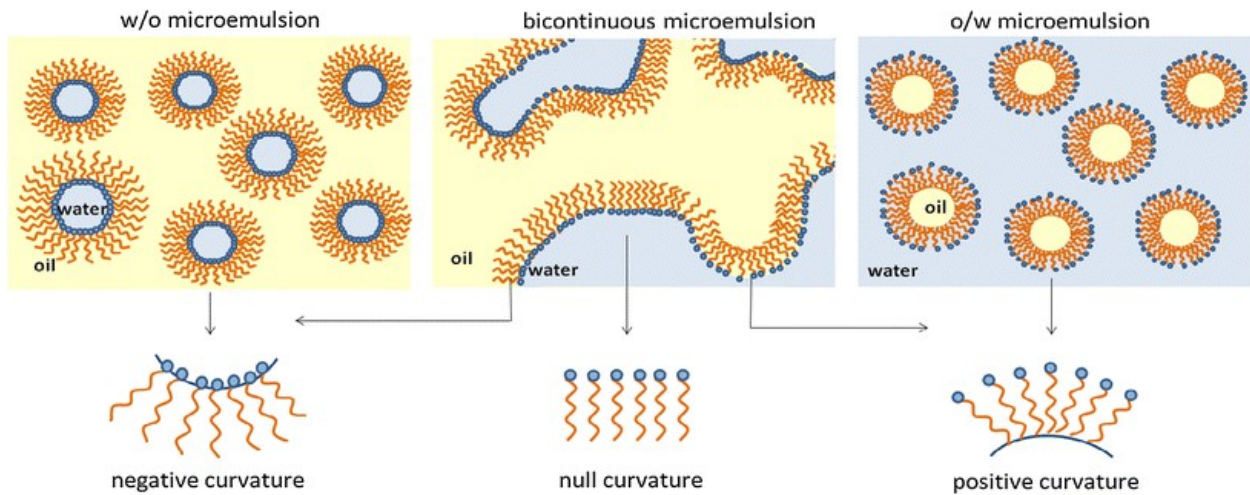


Figure 2. Schematic and monolayer curvature of w/o, o/w and bicontinuous emulsion systems reproduced from reference [11]

Despite their utility, emulsions are intrinsically unstable systems because their formation requires a positive Gibbs free energy. Therefore, long-term persistence is achieved not by true thermodynamic stability, but through kinetic stabilization. Even so, destabilization processes eventually dominate by coalescence, in which droplets merge to reduce surface area; creaming or sedimentation, driven by density differences; and Ostwald ripening, in which small droplets dissolve and redeposit onto larger ones due to Laplace pressure differences [13,14]. To limit destabilization, surfactants act dynamically to reduce interfacial tension and redistribute along the oil–water interface, while solid particles adsorb irreversibly to form rigid interfacial layers that hinder droplet deformation. Emulsion science, therefore, aims to prolong stability to the degree necessary for functional and industrial performance rather than to eliminate instability entirely. [15].

1.2. Industrial Relevance

The ability to disperse immiscible liquids into controlled structures gives emulsions their wide industrial importance across many sectors.

In the pharmaceutical sector, emulsions provide a versatile platform for drug delivery. O/W emulsions are commonly used to solubilize hydrophobic drugs for oral and parenteral administration, while W/O emulsions can encapsulate hydrophilic drugs for controlled release. Nanoemulsions and microemulsions further extend this utility by enabling targeted delivery, enhanced bioavailability, and improved stability of labile therapeutic agents [16–19].

In the petroleum and energy sector, emulsions represent two important aspects of interfacial behavior. O/W emulsions are intentionally employed in enhanced oil recovery (EOR) to increase sweep efficiency and mobilize residual oil. Conversely, W/O emulsions typically emerge during crude extraction and transport, stabilized by surface-active asphaltenes and resins. Although they may provide benefits such as reduced hydrate agglomeration, their persistence poses a major challenge for separation technologies [20,21].

In personal care and cosmetics, emulsions are central to product performance and sensory attributes. Lotions and creams are typically formulated as O/W or W/O emulsions, with the chosen structure determined by the desired texture and skin feel. O/W systems provide lighter textures and faster absorption, whereas W/O systems impart occlusive properties by forming protective films on the skin. These formulations are stabilized by surfactants and polymeric emulsifiers, frequently supplemented with co-surfactants to achieve optimum interfacial balance. In hair care products, emulsions ensure uniform delivery of active ingredients, influencing texture, foaming behavior, and conditioning performance. More broadly, the ability to combine chemically diverse ingredients into stable multiphase systems defines both the sensory qualities and functional performance of cosmetic formulations [22–24].

In food applications, emulsions provide the structural basis for texture, appearance, and flavor release. Milk is an O/W emulsion in which fat droplets are stabilized by proteins and phospholipids within the aqueous phase [25]. In mayonnaise, egg-yolk lecithin and proteins impart interfacial stability and viscosity, producing a dense microstructure that resists phase separation over time

[26]. These examples highlight how emulsion design in foods not only governs texture and mouthfeel, but also determines consumer perception of freshness and quality.

Beyond these core sectors, emulsions are essential to the formulation of paints, coatings, adhesives, agrochemicals, and polymer dispersions. In these applications, droplet size and interfacial chemistry are key formulation variables that shape rheology, application behavior, film formation, and long-term durability of the final product [27–30].

1.3. Stability Challenges: Flocculation, Coalescence, Creaming and Ostwald Ripening

Although emulsions can be kinetically stabilized for extended periods, their inherent thermodynamic instability gives rise to several destabilization pathways, as shown in Figure 3 [31]. The most significant mechanisms are flocculation, coalescence, creaming, and Ostwald ripening, which act individually or in sequence to determine emulsion lifetime.

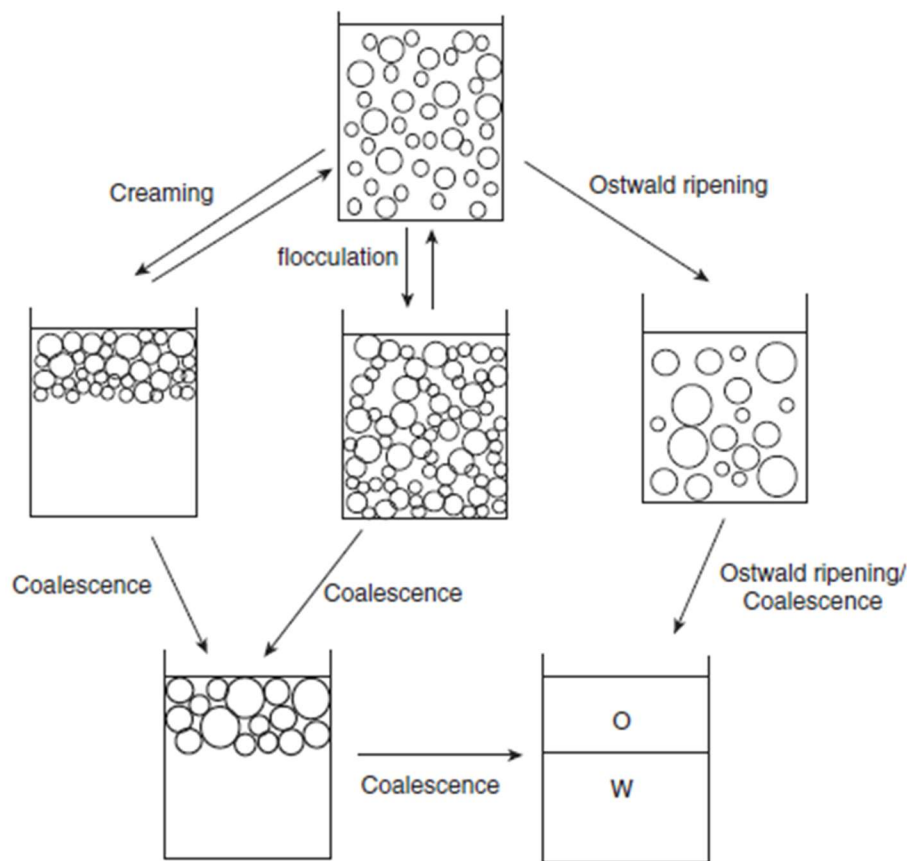


Figure 3. Overview of the key destabilization processes in oil-in-water emulsions and their interrelation reproduced from reference [31]

Flocculation is the reversible association of droplets into clusters while each droplet remains separated by a thin continuous phase film. It occurs when the balance of interparticle forces favors attraction over repulsion. Attractive contributions may include van der Waals interactions, depletion forces caused by non-adsorbing polymers, or bridging by macromolecules that adsorb onto multiple droplet surfaces. Repulsive barriers, typically based on electrostatic or steric effects, determine the degree to which such aggregation proceeds. Flocculation increases the effective

collision frequency of droplets and therefore acts as an important pathway to subsequent destabilization events such as coalescence or gravitational separation. Beyond this role, flocculation also directly modifies emulsion properties by altering apparent droplet size distribution, local concentration, and microstructure. These changes influence viscosity, elasticity, and turbidity, and in some cases promote the formation of extended droplet networks that impart gel-like behavior [32–34].

Coalescence is the irreversible fusion of dispersed droplets, driven by the system's tendency to reduce total interfacial area. It occurs when the thin liquid film between approaching droplets drains to a critical thickness and ruptures, a process that depends strongly on interfacial tension, droplet deformability, and the viscoelastic strength of the interface. As a destabilization pathway, coalescence progressively increases average droplet size and can ultimately lead to phase separation [35,36].

Creaming or sedimentation refers to the gravitational migration of droplets arising from density differences between the dispersed and continuous phases. In O/W emulsions, the dispersed oil droplets are less dense than the aqueous medium and therefore experience a net upward buoyant force that drives them toward the surface. In W/O emulsions, the dispersed aqueous droplets are denser than the continuous oil phase and thus experience a net downward force that causes them to settle. The rate of creaming is predicted by Stokes law, where the velocity scales with the square of the droplet radius, and is reduced by decreasing droplet size or increasing the viscosity of the continuous phase [37–39].

Ostwald ripening is the molecular-scale redistribution of dispersed phase between droplets of different size, driven by differences in solubility and Laplace pressure. Molecules of the dispersed phase diffuse from smaller droplets, which have higher Laplace pressure and greater effective solubility, through the continuous phase and are incorporated into larger droplets, leading to the progressive shrinkage of small droplets and the growth of large ones [40]. Ostwald ripening steadily increases the average droplet size and broadens the size distribution, ultimately reducing long-term stability [41]. The rate of Ostwald ripening can be mitigated by incorporating insoluble oils or colloidal particles that hinder molecular diffusion [42].

These mechanisms constitute the main modes of emulsion destabilization. Formulation strategies are commonly addressed through control of interfacial chemistry, droplet size distribution, and bulk rheology to regulate their extent and impact.

1.4. Surfactant-Stabilized vs. Particle-Stabilized (Pickering) Emulsions

Emulsions are generally stabilized either by molecular surfactants, which adsorb dynamically to the oil–water interface, or by colloidal particles that anchor firmly to the interface, producing Pickering emulsions.

In surfactant-stabilized systems, molecular amphiphiles adsorb rapidly to newly created oil–water interfaces, reducing interfacial tension and enabling droplet breakup under shear [43,44]. Their dynamic adsorption also imparts barriers to coalescence through electrostatic repulsion, steric hindrance, or viscoelastic interfacial films [45]. Because adsorption–desorption is reversible, these interfacial layers remain responsive to changes in concentration, ionic strength, or temperature [46–48]. For instance, amphiphilic molecules migrate from micelles in the aqueous phase to the newly created oil–water interface. The rate and extent of this transfer depend on the surfactant’s solubility, as illustrated in Figure 4, which depicts the dynamic equilibrium between micellar and interfacial adsorption. Note that in Figure 4a, the two colors used for the Tween 60 headgroup represent its two hydrophilic components and do not indicate distinct molecules [49]. This flexibility allows precise control over droplet size and emulsion type (O/W or W/O), but also leaves surfactant-stabilized systems susceptible to long-term instability from molecular exchange and interfacial rearrangement [50].

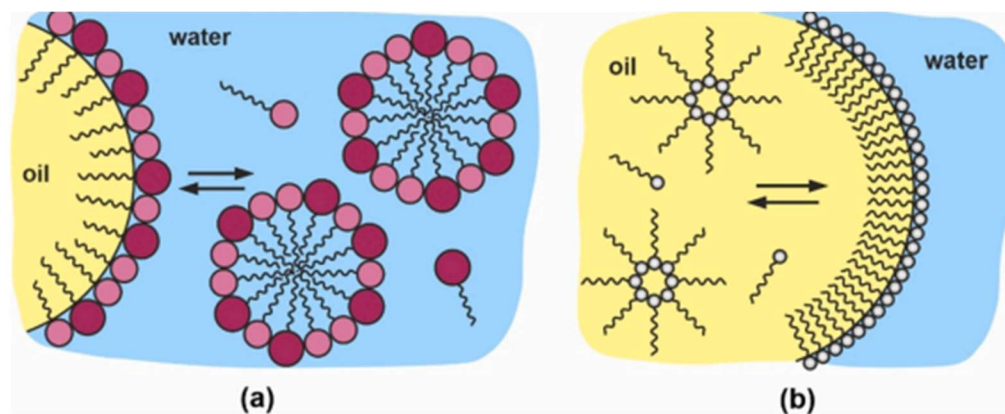


Figure 4. Schematic representation of adsorption layers at the oil–water interface in emulsions stabilized by (a) the water-soluble surfactant Tween 60 and (b) the oil-soluble surfactant Brij 52 reproduced from reference [49].

In Pickering emulsions, colloidal particles that partially wet both phases assemble at the interface into closely packed layers that prevent droplet coalescence. Once in place, particles form rigid,

jammed interfacial structures that impart solid-like rheology and confer exceptional long-term stability, even in the absence of free stabilizers in the continuous phase [51–53], and in some cases, stability can be maintained even at relatively low interfacial coverage [54]. Figure 5 shows a schematic representation of Pickering emulsions, in which droplets are stabilized by solid particles adsorbed at the oil–water interface [55]. The solid particle stabilizers can range from inorganic oxides and metals to polymeric and biomass-derived particles.

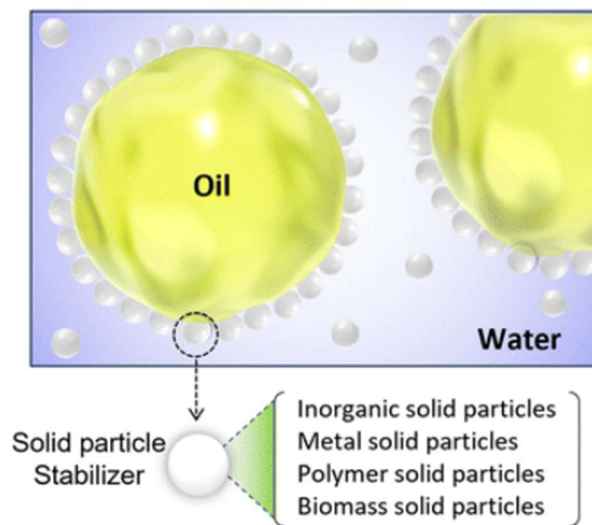


Figure 5. Schematic illustration of Pickering emulsions, where droplets are stabilized by solid particles adsorbed at the oil–water interface reproduced from reference [55]

The limitations of Pickering stabilization include slower interfacial coverage during emulsification and reduced control over droplet size relative to surfactant systems [56]. More details on the mechanisms of particle adsorption and stabilization are discussed in Section 2.3.

1.5. Catastrophic vs. Transitional Phase Inversion

Phase inversion refers to the transformation of an emulsion from one type to the other, characterized by a reversal of continuous and dispersed phases. Two principal pathways are recognized: transitional phase inversion (TPI) and catastrophic phase inversion (CPI). Although both processes lead to a change in phase continuity, the mechanisms and driving forces differ fundamentally.

In transitional phase inversion, the reversal is induced by changes in formulation parameters such as temperature, electrolyte concentration, or surfactant chemistry. For example, nonionic surfactants display a hydrophilic–lipophilic balance (HLB) that shifts with temperature; near the phase inversion temperature (PIT), the surfactant affinity for oil and water becomes balanced, interfacial tension is minimized, and the emulsion reorganizes into the opposite type [57,58]. Likewise, adjustments in salinity, pH, or surfactant ionization state can alter interfacial packing, producing inversion [59,60]. Transitional inversion is thus thermodynamically regulated, strongly dependent on molecular-scale surfactant properties, and is commonly applied in the preparation of microemulsions and nanoemulsions where narrow droplet size distributions are desired [61].

While HLB and PIT concepts are widely used to rationalize such behavior, they are not the only explanatory frameworks. Classical models include Bancroft’s rule, which states that the continuous phase tends to be the one in which the surfactant is more soluble [62]; the critical packing parameter, which links molecular geometry to preferred interfacial curvature [63]; and spontaneous curvature concepts, which emphasize the bending tendency of surfactant monolayers [64]. More recent perspectives include the hydrophilic–lipophilic deviation (HLD) framework, which integrates salinity, temperature, surfactant structure, and oil properties into a predictive parameter [65,66]. In addition, curvature elasticity and free energy landscape approaches, exemplified by the hydrophilic–lipophilic deviation net average curvature (HLD-NAC) model, treat the interface as an elastic film with a preferred spontaneous curvature and predict inversion when formulation variables such as salinity or ionic strength, temperature, oil equivalent alkane carbon number, surfactant headgroup ethoxylation and tail length or branching, and short chain alcohol or cosolvent content shift the free energy minimum from one curvature sign to the other, thereby reversing phase continuity [67,68]. Collectively, these frameworks highlight that

transitional inversion can arise through multiple, system-dependent mechanisms beyond simple HLB shifts. These models are discussed in more detail in section 2.1.

In catastrophic phase inversion, the reversal arises primarily from compositional and hydrodynamic factors rather than interfacial chemistry. Classically, it has been rationalized by phase-volume rules, which propose that an emulsion can only sustain continuity of a given phase up to a critical dispersed-phase fraction. When this threshold is exceeded, the minority phase can no longer form a connected network, and a sudden inversion occurs [69–71]. Early empirical studies established that the critical volume fraction for inversion typically lies between 60–80% [69,72], though the precise value depends strongly on droplet size distribution and mixing conditions [70,71]. These observations indicate that catastrophic inversion is inherently a non-equilibrium process, driven not by molecular-scale interfacial changes but by the inability of the system to preserve continuity of the initial phase under shear.

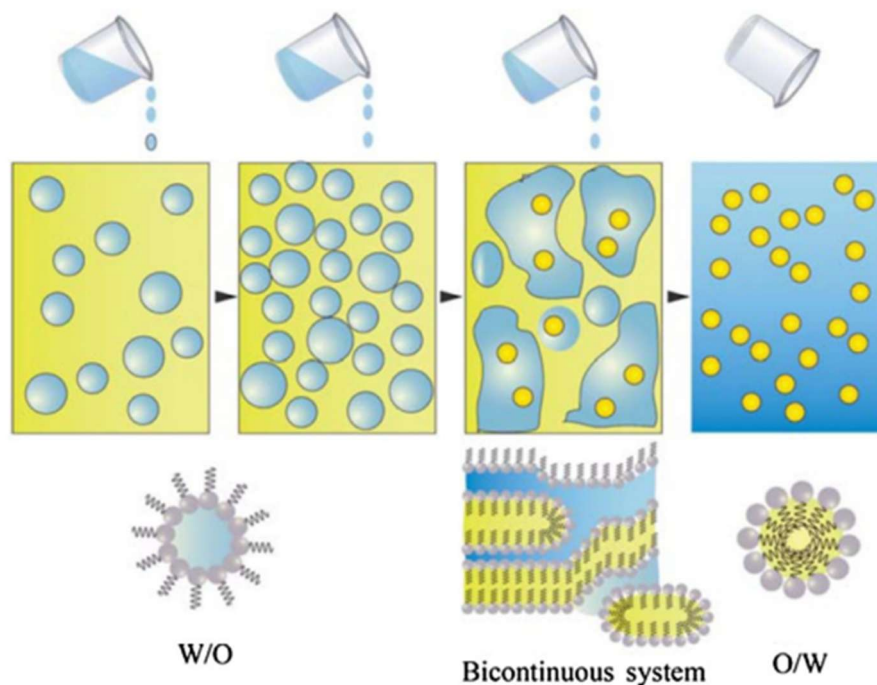


Figure 6. Catastrophic phase inversion (CPI) pathway reproduced from reference [73]

The mechanistic pathway of CPI proceeds through a sequence of interconnected events. As the dispersed phase fraction increases, droplets are forced into close packing, which raises collision frequency and promotes coalescence [70,71]. Under sustained shear, the continuous phase becomes fragmented and can no longer maintain a spanning network, while the dispersed phase

undergoes rapid coalescence events that form a new continuous matrix [70,74]. This restructuring occurs abruptly and yields a broad droplet size distribution, reflecting the disruptive and non-equilibrium character of the transition [70,71]. In contrast to transitional inversion, where ultralow interfacial tension can yield narrow size distributions and precise control, catastrophic inversion is governed by macroscopic flow dynamics and is difficult to reverse once initiated [75].

1.6. Role of Nanocrystal Cellulose (NCC) as Sustainable Stabilizer

The growing demand for renewable formulations with low environmental impact has intensified interest in NCC as stabilizers. NCCs can be produced from forestry and agricultural byproducts, aligning with circular economy and green chemistry principles [76]. Their biodegradability, low toxicity, and biocompatibility make them well-suited to applications where environmental and safety considerations are important, such as food, cosmetics, and biomedical formulations [77–79].

NCCs represent a shift from dynamic molecular surfactants toward colloidal particles that combine sustainability with function. By irreversibly adsorbing at oil–water interfaces, they provide robust droplet stabilization while also influencing rheology and barrier properties [80].

Recent years have seen rising publication trends and commercialization initiatives highlighting the potential of NCCs as sustainable stabilizers. Market analyses forecast strong growth for cellulose nanocrystals, particularly in advanced materials, packaging, coatings, and biomedical applications [81]. The detailed synthesis, properties, and applications of NCCs will be discussed in Section 2.5.

1.7. Knowledge Gaps and Objectives of the Study

Catastrophic phase inversion (CPI) represents a critical yet insufficiently understood transition in emulsion systems. Although both surfactant- and particle-stabilized emulsions have been studied extensively, most investigations consider these regimes separately. As a result, comparative knowledge of how renewable nanomaterials such as nanocrystalline cellulose (NCC) influence CPI relative to conventional surfactants remain scarce.

Surfactants typically promote inversion by reducing interfacial tension through dynamic redistribution, whereas particles such as silica or starch delay inversion by adsorbing irreversibly and producing interfacial crowding. In particular, the ability of NCC, a renewable stabilizer with distinct amphiphilic character and high aspect ratio, to shift inversion thresholds has remained unexplored. To our knowledge, this study provides the first quantitative characterization of catastrophic phase inversion behavior in NCC-stabilized emulsions.

Therefore, the objectives of this study are:

1. **To establish how nanocrystalline cellulose influences the emulsion inversion point (EIP) in catastrophic inversion**, by quantifying shifts in the critical aqueous phase fraction with varying NCC concentration and comparing these effects to those observed with conventional surfactants.
2. **To relate interfacial properties to inversion behavior**, through surface and interfacial tension measurements that distinguish between dynamic surfactant-driven mechanisms and particle-based steric or jamming effects.
3. **To assess the stability of water-in-oil emulsions prior to inversion**, by measuring coalescence resistance and identifying the concentration ranges where NCC or surfactants provide the most effective stabilization.

The objectives outlined here provide the framework for the work that follows. The overall structure of the thesis is presented in the next section.

1.8. Thesis Structure

This thesis is organized into six chapters.

- **Chapter 1** provides an introduction, outlining the background, motivation, and objectives of the research.
- **Chapter 2** reviews relevant literature on emulsion stability, phase inversion mechanisms, surfactant- and particle-stabilized systems, and the emerging role of nanocellulose as a functional stabilizer.
- **Chapter 3** describes the materials, preparation methods, and experimental techniques used to investigate phase inversion and emulsion stability.
- **Chapter 4** presents the results of the experimental investigations, with emphasis on phase inversion behavior, coalescence stability, and interfacial dynamics.
- **Chapter 5** discusses the results in relation to theoretical frameworks and prior studies, highlighting the mechanistic implications of NCC on interfacial dynamics and inversion processes.
- **Chapter 6** summarizes the conclusions and outlines future research directions.

Chapter 2. – Literature Review

2.1. Theoretical Foundation of Emulsions

The stability of emulsions reflects a balance between the thermodynamic drive toward phase separation and the kinetic barriers created at liquid–liquid interfaces. This framework explains the conditions under which emulsions form, the structures they adopt, and the mechanisms by which they resist destabilization.

2.1.1. Thermodynamics of Dispersion

Dispersing one liquid into another creates a substantial increase in interfacial area, and hence in interfacial free energy. The Gibbs free energy of emulsification can be expressed as:

$$\Delta G = \gamma \Delta A - T \Delta S \quad (1)$$

where γ is the oil–water interfacial tension, ΔA is the newly created surface area, and ΔS accounts for the entropy increase associated with droplet formation [15]. Because γ is typically large (10–50 mN·m⁻¹), the positive $\gamma \Delta A$ term dominates, making emulsions thermodynamically unstable. Emulsification thus requires energy input (e.g., shear, homogenization) to overcome this barrier. Emulsion systems persist only when kinetic stabilization mechanisms such as electrostatic repulsion, steric barriers, or rigid interfacial films elevate the energy barrier to droplet coalescence [82,83].

2.1.2. Interfacial Tension and Film Formation

Surfactants lower interfacial tension by adsorbing at the oil–water interface, reducing the energy cost of creating new surface. This relationship is described by the Gibbs adsorption equation:

$$d\gamma = - \sum_i \Gamma_i d\mu_i \quad (2)$$

Where Γ_i is the surface excess concentration of component i , and μ_i is its chemical potential. The equation shows that interfacial tension decreases as surfactant molecules accumulate at the interface [82].

At sufficient coverage, surfactants assemble into interfacial films that act as dynamic barriers to droplet rupture and coalescence, with viscoelastic properties that can be quantified using interfacial rheology. When two droplets approach, DLVO theory explains the balance of forces, with van der Waals attraction driving film thinning while electrostatic repulsion opposes coalescence [83,84]. Additionally, interfacial stability also depends on dynamic processes such as the Gibbs–Marangoni effect. When a droplet interface is perturbed, surfactants redistribute unevenly, creating local tension gradients that drive restorative flows back toward depleted regions. This self-healing response counteracts thinning and rupture, helping emulsions withstand collisions and shear where purely electrostatic forces might fail [82,83,85].

Collectively, these mechanisms contribute to the stabilization of the oil–water interface and allow surfactants to preserve emulsions as metastable systems.

2.1.3. HLB, HLD, HLD-NAC Frameworks

HLB

The hydrophilic–lipophilic balance (HLB) scale provides a foundational framework for surfactant classification. In Griffin’s formulation, developed for nonionic surfactants, the HLB value is derived from the ratio of the molecular mass of the hydrophilic portion (M_h) to the total molecular mass (M) [86]:

$$HLB = 20 \times \frac{M_h}{M} \quad (3)$$

Davies later refined this approach by introducing a group contribution method, in which numerical constants are assigned to specific hydrophilic (H_i) and lipophilic (L_j) groups [87]:

$$HLB = 7 + \sum H_i - 0.475 \sum L_j \quad (4)$$

This refinement extended the framework to ionic and more structurally complex surfactants, providing broader classification [87].

Characteristic HLB ranges link to functionality: low values ($\approx 3\text{--}6$) favor water-in-oil emulsions, high values ($\approx 8\text{--}18$) favor oil-in-water systems, and intermediate values support wetting and dispersion [86–88].

Despite its simplicity and continued industrial use, the HLB framework has limited predictive accuracy when applied across systems with varying oil polarity, electrolyte content, or temperature.

HLD

To overcome the simplified assumptions of the HLB concept, the hydrophilic lipophilic difference (HLD) framework was introduced as a quantitative tool to better predict emulsion type and phase behavior. It incorporates key formulation variables such as oil composition, salinity, surfactant structure, temperature, and cosurfactant content into a dimensionless balance equation that represents the overall affinity of a surfactant for oil and water phases. The framework enables predictive understanding of emulsion type, phase transitions, and interfacial properties using measurable physicochemical variables.

The general form of HLD equation is expressed as [89,90]:

$$HLD = +C_c - k \cdot EACN - \alpha \Delta T + f(S) \quad (5)$$

where C_c is the characteristic curvature of the surfactant that represents its intrinsic hydrophilic or lipophilic tendency, $EACN$ (equivalent alkane carbon number) characterizes the oil phase, ΔT is the deviation from the reference temperature, and $f(S)$ accounts for the effect of salinity or ionic strength. Constants k and α are empirical coefficients that depend on the surfactant class [89,90].

The sign and magnitude of HLD describe how formulation variables collectively shift the interfacial balance between oil and water affinity. When $HLD = 0$, the system reaches an optimum formulation where the surfactant exhibits equal chemical potential in both phases. At this balanced condition, the interfacial curvature approaches zero, interfacial tension reaches its minimum, and solubilization of both oil and water phases is maximized. Negative values ($HLD < 0$) correspond to hydrophilic conditions that favor oil in water structures, whereas positive values ($HLD > 0$) indicate lipophilic conditions that favor water in oil structures [90–92].

The conceptual distribution of emulsion types across the HLD scale is illustrated in Figure 7, which shows how variations in formulation balance govern the transition between oil-in-water and water-in-oil regimes. Near the balanced region ($HLD \approx 0$), the interfacial energy is finely tuned, allowing emulsions to form easily under low shear. These fine emulsions arise where the curvature is most flexible and the interfacial tension lowest, though they are generally short-lived due to the transient nature of the balance [90,93].

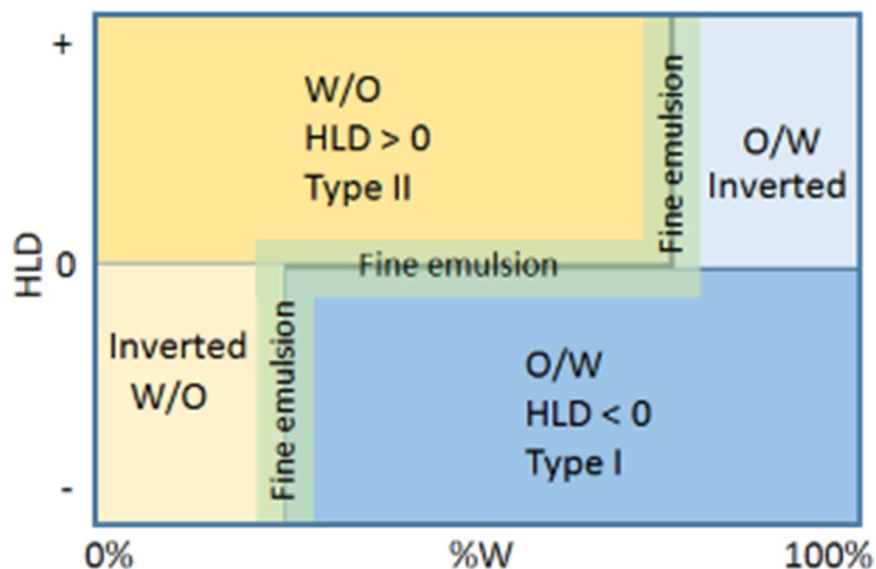


Figure 7. Schematic representation of the hydrophilic lipophilic difference (HLD) scale showing phase behavior and emulsion type as a function of formulation balance [90,93]

Each term in the HLD equation reflects a distinct physical contribution to the interfacial balance. Salinity increases the HLD value because added salt screens electrostatic repulsion and reduces hydration of ionic surfactant headgroups, shifting them toward the oil phase. The salinity term $f(S)$ depends strongly on surfactant type: it is approximately $0.13 \times S$ for ethoxylates, nearly constant for nonionic surfactants such as sorbitan esters and alkyl polyglucosides (APGs), and proportional to $\ln(S + \text{SurfSal})$ for ionic surfactants. Here, SurfSal represents the intrinsic salinity contribution from the ionic surfactant itself, which releases counterions into solution and can be approximated as about 30 percent of its molar NaCl equivalent. Increasing the oil phase hydrophobicity, expressed by a larger $EACN$, proportionally decreases the HLD value, with k ranging from about 0.15 to 0.17 for most surfactants and near 0.06 for extended surfactants that contain flexible polyoxypropylene linkers and are therefore less sensitive to oil polarity.

Temperature also modulates the formulation balance through the temperature coefficient (α), which quantifies the thermal sensitivity of surfactant hydration. A rise in temperature slightly lowers HLD for ionic surfactants ($\alpha \approx 0.01$), elevates HLD for ethoxylates due to dehydration of the ethoxy chains ($\alpha \approx -0.06$), and exerts negligible influence for alkyl polyglucosides ($\alpha \approx 0$). The characteristic curvature (C_c) term adjusts the overall balance according to the intrinsic hydrophilic–lipophilic nature of the surfactant, with more positive values corresponding to stronger affinity for the oil phase and therefore higher HLD values [90].

Experimental validation has consistently demonstrated the robustness of the HLD framework across diverse surfactant classes. Schirone et al. (2021) expanded the model to cationic and amphoteric surfactants, showing that the characteristic curvature correlates linearly with molecular hydrophobicity and aligns with conventional HLB values [89]. Zheng et al. (2022) examined mixed ionic–nonionic systems comprising Span-80 and sodium laureth sulfate, demonstrating that formulations with mixed HLD values between -1 and $+1$ undergo spontaneous curvature fluctuations and self-emulsification under ultralow interfacial tension, confirming the model’s ability to capture both equilibrium and dynamic interfacial behavior [94]. Wang et al. (2024) further validated HLD relationships in nonionic block polyether surfactants, establishing proportionality among the hydrophobicity constant (k), temperature coefficient (α), and characteristic curvature (C_c), and confirming that variations in EO/PO ratio and molecular configuration systematically influence the apparent HLD response[95]. Together, these studies affirm that the HLD framework captures systematic trends in surfactant behavior across ionic, nonionic, amphoteric, and mixed systems.

From a thermodynamic perspective, HLD expresses the difference in surfactant chemical potential between the oil and aqueous phases, providing a quantitative description of formulation balance. Its strength lies in translating molecular interactions into a measurable descriptor of interfacial behavior, forming the foundation for subsequent models such as HLD-NAC that extend this balance to geometric and curvature-based interpretations.

HLD-NAC

As previously outlined, the HLD framework quantifies formulation balance by describing how changes in variables such as salinity, temperature, and surfactant structure shift a system's affinity between oil and water. However, it remains an equilibrium descriptor, and it does not describe how the interface deforms or resists bending when displaced from that balanced state.

To overcome these limitations, Acosta and coworkers [96] developed the hydrophilic lipophilic deviation net average curvature (HLD-NAC) model. This model integrates the compositional insight of HLD with a geometric description of the interface, translating formulation parameters into physically measurable quantities such as curvature, solubilization, and interfacial tension. The central idea is that the NAC model treats the interface between oil and water as a deformable surface whose curvature adjusts according to the relative chemical potential of surfactant molecules in each phase [96].

The net curvature (H_n) provides a quantitative translation of the HLD value into a geometric property of the interface:

$$H_n = -\frac{\text{HLD}}{L} \quad (6)$$

where L is the characteristic surfactant length, corresponding to the extended tail length of the surfactant molecule [96].

A positive H_n indicates that the interface bends toward water, forming oil dispersed in water structures, while a negative H_n reflects curvature toward oil, forming water dispersed in oil systems. At $H_n = 0$, the interface attains zero mean curvature, corresponding to the balanced state where the surfactant has equal chemical potential in both phases and the interfacial tension reaches its minimum.

The average curvature ($H_a = 1/\xi$) describes how tightly or loosely the surfactant film is curved on average [96]. It is determined by the correlation length (ξ), which represents the distance over which the interface remains smoothly connected before its curvature changes direction. In physical terms, ξ corresponds to the typical spacing between adjacent oil and water domains separated by the surfactant layer. Small ξ values indicate a tightly curved interface with fine domains, while large values correspond to smoother curvature and broader, more continuous structures.

The interfacial rigidity (E_r) measures how strongly the surfactant film resists bending. A film with high rigidity requires more energy to curve, while a flexible film bends easily under small changes in formulation or stress [96]. This property determines how stable the interface remains when the system moves away from the balanced condition. Interfacial rigidity is directly related to the interfacial tension ($\gamma = E_r/4\pi R^2$), linking the molecular elasticity of the surfactant layer to measurable macroscopic behavior such as droplet deformation, coalescence resistance, and overall emulsion stability [96].

The parameters H_n , H_a , and E_r link formulation variables to interfacial curvature, structure, and elasticity. Their interplay extends the HLD concept into a predictive model that describes how the interface softens and reorganizes near balance, offering deeper insight into emulsion inversion behavior.

In practice, the hydrophilic lipophilic deviation–net average curvature (HLD–NAC) model has moved beyond theory to become a reliable formulation tool used across cosmetic, industrial, and environmental systems. In cosmetic and personal-care formulations, it has been applied to optimize the interfacial curvature and solubilization behavior of surfactant mixtures used in skin cleansers, moisturizers, and fragrance emulsions [97].

In industrial demulsification and wastewater treatment, the HLD–NAC model has been used to predict and optimize oil–water separation. Ghasemi and Eslami [98] showed that model-derived parameters such as surfactant length and droplet aspect ratio correlated strongly with experimental demulsification efficiency in petrochemical wastewater. Formulations exhibiting higher curvature anisotropy achieved faster and more complete oil removal, confirming that the geometric descriptors of the HLD–NAC framework can reliably indicate separation performance and guide demulsifier design [98].

2.2. Surfactant Chemistry and Stabilization Mechanisms

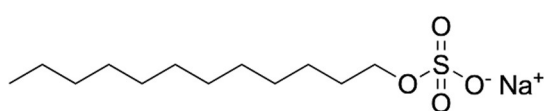
Surfactants are amphiphilic molecules that adsorb at oil–water interfaces, where they lower interfacial tension and promote droplet formation while inhibiting coalescence. Their molecular architecture comprises a hydrophobic tail and a hydrophilic headgroup, and the chemical nature of these components governs interfacial interactions and the dominant stabilization mechanism [15]. Based on headgroup functionality, surfactants are generally classified as anionic, cationic, nonionic, or zwitterionic, each characterized by distinct interfacial behavior and physicochemical properties.

Anionic surfactants, such as *sodium dodecyl sulfate (SDS)*, consist of a dodecyl hydrocarbon tail and a sulfate headgroup that dissociates to yield negatively charged ions in aqueous solution. The resulting surface charge generates electrostatic repulsion between droplets and forms a diffuse electrical double layer that suppresses coalescence [99]. SDS is one of the most extensively studied model surfactants and is widely used as a benchmark for interfacial and micellization studies due to its well-defined CMC and predictable adsorption behavior at oil–water interfaces [100,101].

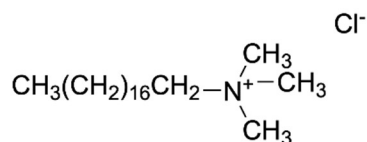
Cationic surfactants, exemplified by *octadecyltrimethylammonium chloride (OTAC)*, feature a long C₁₈ alkyl chain attached to a quaternary ammonium headgroup. When adsorbed at the interface, OTAC can impart a net positive charge to the droplet surface, enhancing stability through electrostatic repulsion among droplets. The long hydrocarbon tail promotes strong adsorption and compact interfacial packing, though chloride counterions can influence charge screening and micelle morphology [102]. For example, micelles of OTAC have been characterized in terms of size and structure in aqueous solution [103]. OTAC is used across a wide range of applications such as crude oil flow improvement and corrosion inhibition [104,105]

Nonionic surfactants, such as *C_{12–14} alcohol ethoxylate (Alfonic 1412-3)*, are composed of mixed-chain linear fatty alcohols with three ethylene oxide (EO) units that provide hydrophilicity through hydrogen bonding with water. Stabilization occurs predominantly through steric effects generated by hydrated ethylene oxide chains that hinder droplet coalescence [106,107]. Because they lack ionic charge, Alfonic surfactants show minimal sensitivity to electrolyte concentration and retain interfacial activity across a wide pH range, making them suitable for diverse industrial and oilfield applications [1].

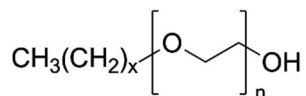
Zwitterionic surfactants, including *cetyl betaine* (*Amphosol*), contain both a quaternary ammonium cation and a carboxylate anion within the same molecule, typically linked through a short alkyl spacer. This dual functionality allows charge adaptation with pH and ionic strength, enabling stabilization through both electrostatic and steric effects [108,109]. Cetyl betaine is valued for its mild interfacial behavior and compatibility with a wide range of surfactants. It is often employed as a co-surfactant to adjust interfacial curvature and enhance formulation stability under varying salinity and pH conditions. [110].



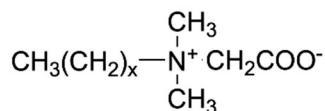
Sodium dodecyl sulfate (SDS)



Octadecyltriethylammonium chloride (OTAC)



Alcohol ethoxylate; x=12-14, n=3



Cetyl betaine; x=12-16

Figure 8. Representative molecular structures of surfactants used in this study. (a) Sodium dodecyl sulfate (SDS, anionic); (b) Octadecyltrimethylammonium chloride (OTAC, cationic); (c) C₁₂₋₁₄ alcohol ethoxylate, (nonionic); and (d) Cetyl betaine (zwitterionic)

The effectiveness of surfactants in stabilizing emulsions depends strongly on their bulk concentration and adsorption behavior at the interface. At concentrations below the critical micelle concentration (CMC), surfactant molecules preferentially migrate to the oil–water interface, progressively lowering interfacial tension and facilitating droplet breakup during emulsification [1,15].

Once the CMC is reached, additional molecules self-assemble into micelles within the aqueous phase, altering the partitioning of surfactant species between the interface and bulk solution. For ionic surfactants such as SDS and OTAC, micellization is influenced by electrostatic interactions and counterion binding [111].

In contrast, nonionic surfactants like alcohol ethoxylates exhibit lower CMC values due to the absence of electrostatic repulsion, and their adsorption is driven primarily by hydrogen bonding and entropic stabilization of hydrated ethylene oxide chains [112].

Zwitterionic surfactants such as Amphosol display intermediate behavior, as their headgroups can interact through both electrostatic attraction and dipolar association, resulting in flexible adsorption and curvature adjustment near the interface [113,114].

Surfactant-stabilized emulsions rely on dynamic adsorption and interfacial tension reduction to maintain stability. These mechanisms differ fundamentally from those of particle-stabilized systems, where interfacial adsorption is irreversible and governed by particle geometry and surface chemistry, as discussed in the next section.

2.3. Particle-Stabilized (Pickering) Emulsion

In contrast to surfactant-stabilized systems, particle-stabilized emulsions, commonly referred to as *Pickering emulsions*, exhibit markedly different inversion behavior and interfacial dynamics. In these systems, stability and phase inversion are governed not by molecular adsorption–desorption equilibria but by the irreversible adsorption of colloidal particles at the oil–water interface [115,116]. The adsorption of particles substantially reduces interfacial energy, and once attached, the energy required for desorption typically exceeds several thousand times the thermal energy (kT) [117,118]. As a result, the interfacial film becomes quasi-permanent, providing long-term stability against coalescence and Ostwald ripening [115,118].

For a spherical particle of radius R and contact angle θ at an oil–water interface with interfacial tension γ_{ow} , the desorption energy is

$$\Delta E = \pi R^2 \gamma_{ow} (1 \pm \cos\theta)^2 \quad (7)$$

where θ is the contact angle measured through the water phase. The minus sign corresponds to detachment into the water phase and the plus sign to detachment into the oil phase. For nanoparticles with intermediate wettability, the desorption energy exceeds the thermal energy by orders of magnitude, which is a key contributor to the exceptional stability of Pickering emulsions [119].

At high surface coverage, the interfacial layer enters a jammed, solid like state that restricts droplet deformation and suppresses coarsening [120]. The jammed interfacial network limits coarsening pathways and raises the dispersed phase volume needed for inversion, consistent with limited coalescence in solid stabilized emulsions [121,122].

Particle shape and aspect ratio dictate how particles pack and bear stress at the interface. Anisotropic particles often generate stronger interfacial rigidity and higher effective coverage than spheres, which enhances resistance to coalescence and can shift inversion thresholds [123,124].

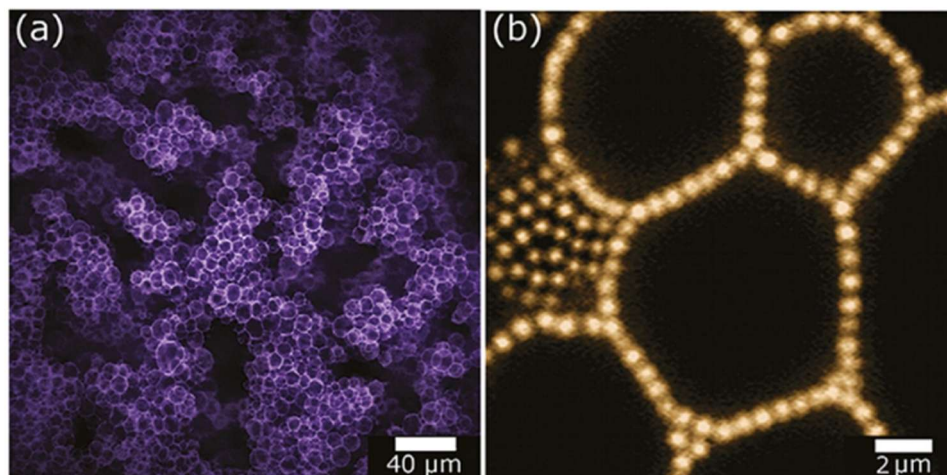


Figure 9. Confocal microscopy images of Pickering emulsion gels. (A) Low-magnification view showing interconnected droplet clusters. (B) High-magnification view revealing closely packed particles bridging adjacent droplets in the gel network. Images from reference [125]

Wettability dictates emulsion type through the particle contact angle at the oil water interface. Particles with contact angles below 90° preferentially stabilize oil in water, while those above 90° favor water in oil; the phase that wets the particle more strongly tends to become the continuous phase [115,116].

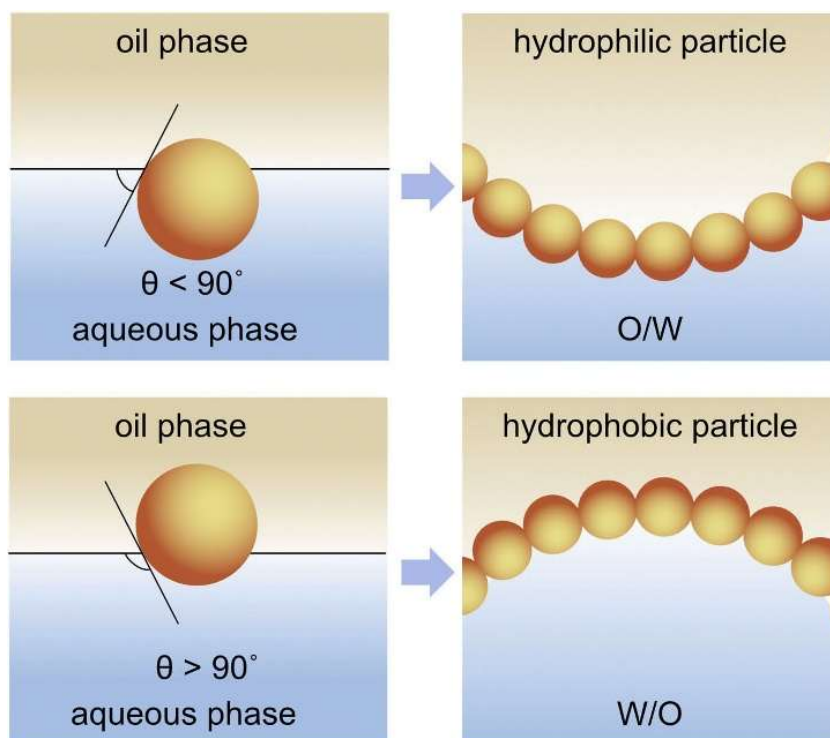


Figure 10. Schematic of pickering emulsion and effect of particle angle on interface curvature [123]

Biobased colloids such as cellulose, starch, chitosan, lignin, and plant proteins have gained attention as Pickering stabilizers because they are renewable and scalable, allow straightforward surface modification to tune wettability, and form robust interfacial layers that suppress coalescence under practical processing conditions[116,123]

Overall, particle stabilized emulsions are governed by a mechanically protected interface whose behavior is set by wettability, coverage, and particle architecture rather than by rapid adsorption and desorption. These features provide the context for catastrophic inversion, where transitions are driven by particle or surfactant stabilization mechanisms that can delay or redirect the inversion pathway.

2.4. Catastrophic Phase Inversion

Catastrophic phase inversion (CPI) refers to the abrupt reversal of emulsion type, typically from oil in water to water in oil or vice versa, caused by changes in phase volume ratio or flow conditions rather than by variations in surfactant affinity or formulation balance, as shown in Figure 11. Unlike transitional inversion, which occurs near the balanced formulation predicted by the hydrophilic lipophilic deviation ($HLD = 0$), CPI arises under nonequilibrium conditions where hydrodynamic instabilities, droplet crowding, and coalescence collectively alter phase continuity. It is therefore governed primarily by flow induced deformation and coalescence dynamics rather than equilibrium interfacial curvature or surfactant chemical potential [126].

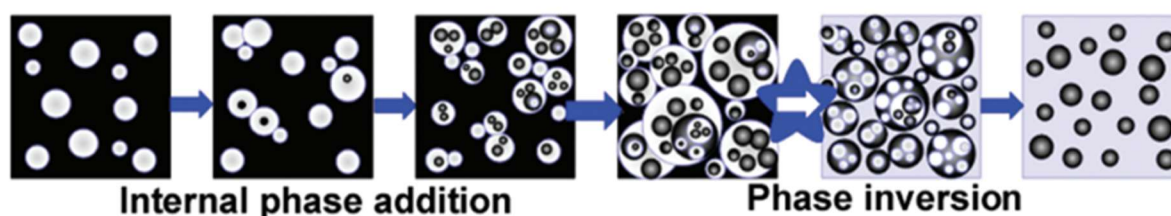


Figure 11. Schematic of catastrophic phase inversion reproduced from reference [127]

The concept of catastrophic phase inversion was first identified by Shinoda and Saito [128], who observed that emulsions can invert even when interfacial tension still favors the original phase, provided the dispersed phase fraction exceeds a critical limit [128]. As the dispersed phase fraction increases, droplets become densely packed and interact more frequently, leading to deformation and partial coalescence. [126,129].

Figure 12 shows a two-dimensional formulation–composition map at constant surfactant concentration illustrates how emulsion type varies with formulation parameters and oil–water ratio. Regions B, A, and C correspond respectively to oil-rich, intermediate, and water-rich phases, while the “+” and “–” symbols indicate the sign of the hydrophilic–lipophilic deviation (HLD). The bold line, known as the inversion frontier, separates water-in-oil (W/O) from oil-in-water (O/W) regions. Crossing this line horizontally (by varying composition) corresponds to CPI, whereas crossing vertically (by changing formulation variables) represents transitional phase inversion (TPI). In practice, inversion is not instantaneous but may be delayed due to the formation of multiple emulsions, leading to triangular uncertainty zones where the exact boundary is indistinct. These

regions account for the observed hysteresis, where the emulsion structure is not fully reversible upon cycling the water-to-oil ratio [73].

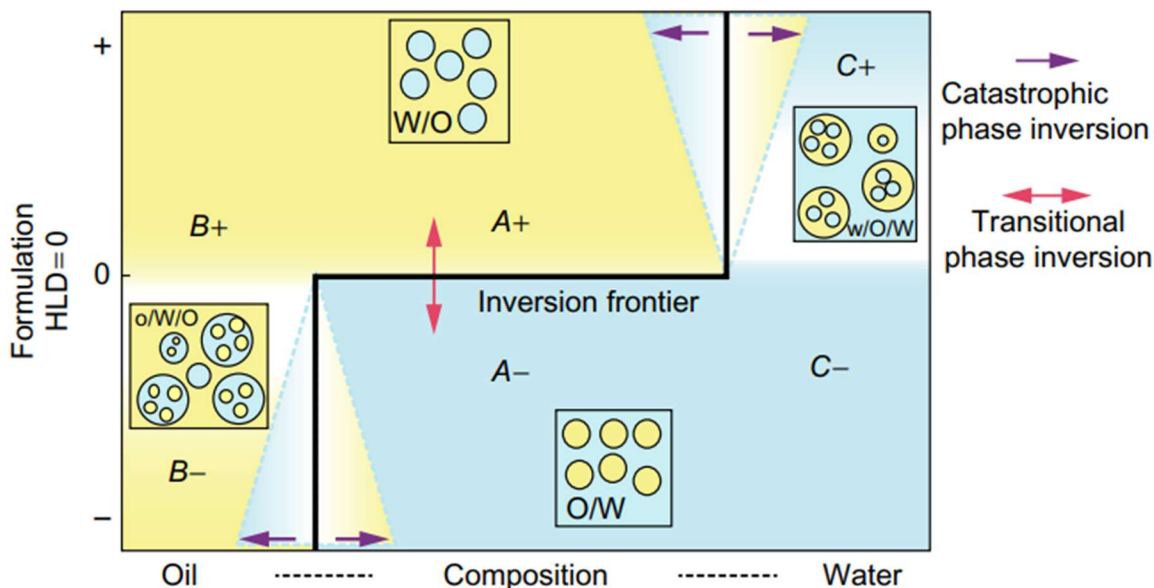


Figure 12. Formulation–composition diagram at a fixed surfactant concentration. The bold line marks the phase inversion boundary. The water-to-oil ratio changes along the horizontal axis of the plot. Reproduced from reference [127]

CPI is characterized by abrupt changes in measurable properties such as viscosity, electrical conductivity, torque, and visual appearance. During inversion, viscosity reaches a distinct maximum due to droplet crowding and temporary bicontinuity, then drops sharply once the new phase network forms [126,130]. Conductivity also changes suddenly as the continuous phase shifts between water and oil, providing a clear marker of the inversion point [121].

Theoretical descriptions of phase inversion have evolved from simple geometric and thermodynamic models to more sophisticated kinetic frameworks that account for dynamic restructuring under flow. Early attempts by Ostwald proposed that the attainable dispersed-phase volume fraction could not exceed the close-packing limit of hard spheres (≈ 0.74 for monodisperse droplets) [131], but could not describe system's composition or processing conditions.

Thermodynamic models based on catastrophe theory describe phase inversion as a transition on a free energy surface, where the system moves smoothly between oil in water and water in oil states through continuous and reversible pathways. This framework captures the equilibrium and

hysteresis-free behavior typical of transitional inversion but cannot represent the abrupt and irreversible changes that occur under the nonequilibrium mixing and coalescence conditions characteristic of catastrophic inversion [132].

Earlier kinetic interpretations, such as that of Vaessen [133] and co-workers, described catastrophic inversion as arising when coalescence rates exceed droplet breakup rates under increasing shear or dispersed-phase content [133]. More recent studies have expanded this view by linking inversion to local energy dissipation, transient bicontinuity, and flow-induced structural transitions [126,134,135].

2.4.1. Phase Inversions in Surfactant-Stabilized Systems

In emulsions stabilized by molecular surfactants, inversion arises from the dynamic redistribution of surfactant molecules between interfaces and bulk phases as composition or flow conditions change. Because surfactant adsorption and desorption occur on short timescales, the interfacial film remains mobile and can readily reorganize under shear. As the dispersed phase fraction increases, droplet collisions intensify and thin films separating them drain rapidly, accelerating coalescence once interfacial tension gradients can no longer be balanced by surfactant diffusion. This process culminates at a critical composition known as the emulsion inversion point (EIP), where phase continuity reverses abruptly [1,132].

This mechanism has been supported by conductivity and light scattering studies that track the EIP and show abrupt increases in droplet size and polydispersity near the inversion threshold [10,136]

Furthermore, studies have shown that phase inversion is accompanied by viscosity divergence and transient formation of multiple emulsions, consistent with kinetic models involving shear-induced instability and droplet–droplet interactions [137,138]. These findings collectively support the view that catastrophic inversion is a nonequilibrium process, proceeding through metastable intermediates rather than a single thermodynamic transition.

2.4.2. Phase Inversions in Particle-Stabilized Systems.

In these systems, phase inversion and overall emulsion stability are governed primarily by the wetting properties, size, and surface coverage of the particles at the oil–water interface. Because particle adsorption is effectively irreversible, the interfacial layer formed is mechanically rigid, which alters both the dynamics and the onset of inversion [139,140].

Due to the irreversible adsorption and mechanical rigidity of the particle-laden interface, phase inversion can be significantly delayed or even suppressed when particles form a densely packed or jammed interfacial layer [141]. This resistance to inversion arises because the particle-stabilized droplets become less deformable and less prone to coalescence, effectively raising the critical internal phase volume required for structural transition. Moreover, the inversion point in Pickering systems can be further shifted by modifying particle hydrophobicity, aspect ratio, or interfacial elasticity, highlighting the importance of particle–interface interactions in controlling emulsion morphology [142,143].

As a result, the catastrophic phase inversion of Pickering emulsions is typically delayed to significantly higher dispersed phase volume fractions. This behavior has been demonstrated in several systems using different types of nanoparticles. For instance, Bains and Pal [121] showed that starch nanoparticles (SNPs) stabilized W/O emulsions and shifted the inversion point to higher aqueous volume fractions, correlating the delay with enhanced nanoparticle concentration and interfacial packing density [121]. Similarly, Malhotra and Pal [144] observed that hydrophobic silica nanoparticles increased the phase inversion point concentration compared to hydrophilic ones, further emphasizing the role of wettability and particle concentration in delaying inversion [144]. Ogunlaja et al. [145] reported that both hydrophilic and hydrophobic starch nanoparticles remained adsorbed at the interface and contributed to increased emulsion stability and delayed inversion points. Furthermore, combining nanoparticles with surfactants to form hybrid stabilizers has been shown to synergistically enhance emulsion stability while extending the inversion point beyond what is achievable with either component alone [145].

This is consistent with the findings from Binks et al. [69], who demonstrated that hydrophobic silica nanoparticles delayed inversion of W/O emulsions to O/W emulsions until water volume fractions approached ≈ 0.7 , far exceeding typical inversion points seen with surfactants alone [69]. Further, Zanini et al. [135] reported that graphene oxide and silica-coated particles exhibited

delayed phase inversion due to the formation of rigid interfaces that resisted droplet coalescence. It was also shown that the surface roughness of colloids can induce contact angle hysteresis, leading to enhanced emulsion stability and resistance to inversion under mechanical agitation [135]. Similarly, González-Cruz et al. [146] investigated the effects of pH and ionic strength on Pickering emulsions stabilized by palmitic acid–decorated silica nanoparticles. Their findings indicated that the stability limit and inversion threshold are controlled by the degree of particle ionization, with higher surface charge resulting in a more robust interfacial layer, thereby delaying phase inversion and enhancing emulsion stability [146].

These insights highlight the broader importance of Pickering emulsions as robust, particle-stabilized systems that can maintain emulsion stability under demanding conditions. This is particularly relevant in energy applications, such as enhanced oil recovery, where stable Pickering emulsions have been shown to improve oil displacement efficiency and provide effective flow diversion and conformance control in high-temperature, high-salinity reservoirs [147].

2.5. Nanocrystalline Cellulose (NCC)

Nanocrystalline Cellulose (NCCs) have emerged as a renewable class of nanomaterials distinguished by their highly ordered crystalline structure and versatile surface chemistry. Derived from cellulose, the most abundant biopolymer in nature, NCCs exhibit high crystallinity, mechanical rigidity, and adjustable interfacial chemistry, making them valuable in a wide range of colloidal, coating, and biomedical applications.

2.5.1. Synthesis

NCCs are typically obtained through controlled depolymerisation of lignocellulosic biomass, where the amorphous portions of cellulose are hydrolyzed to isolate the crystalline fraction [148].

Mineral acid hydrolysis remains the predominant approach: sulfuric acid cleaves amorphous cellulose and installs sulfate half ester groups that generate negative surface charge and good aqueous dispersibility, whereas hydrochloric or phosphoric acid yields largely uncharged nanocrystals that display higher thermal stability but disperse less readily in water. These outcomes and their dependence on reaction conditions are well established in recent reviews and benchmark studies [149–151].

Beyond conventional acid hydrolysis, selective oxidation and alternative solvent systems have been developed to tailor the surface chemistry of NCCs. TEMPO-mediated oxidation converts primary hydroxyl groups at the C6 position into carboxylate groups under mild aqueous conditions, producing negatively charged nanocrystals with improved colloidal stability. This method can also be combined with acid hydrolysis to enhance re-dispersibility, and recent variations use electrochemical TEMPO systems together with HCl gas hydrolysis to reduce reagent use [152,153].

Ionic liquid processing, particularly with imidazolium-based solvents, can partially dissolve cellulose and regenerate nanocrystals with controllable dimensions and surface characteristics, offering a tunable and recyclable route to NCC preparation [148,154].

2.5.2. Structural and Surface Properties

The morphology and physical characteristics of NCCs depend strongly on the cellulose source and the conditions used during hydrolysis. The nanocrystals are generally rod-shaped, with widths ranging from 5 to 20 nm and lengths between 100 and 500 nm, giving typical aspect ratios of 10–50 [148,155].

Presented by the Pal group [156], Figure 13 shows the SEM images of the agglomerated NCC networks, TEM and AFM images of individual NCC crystals which clearly show that the nanocrystals are rod- or needle-shaped particles [156].

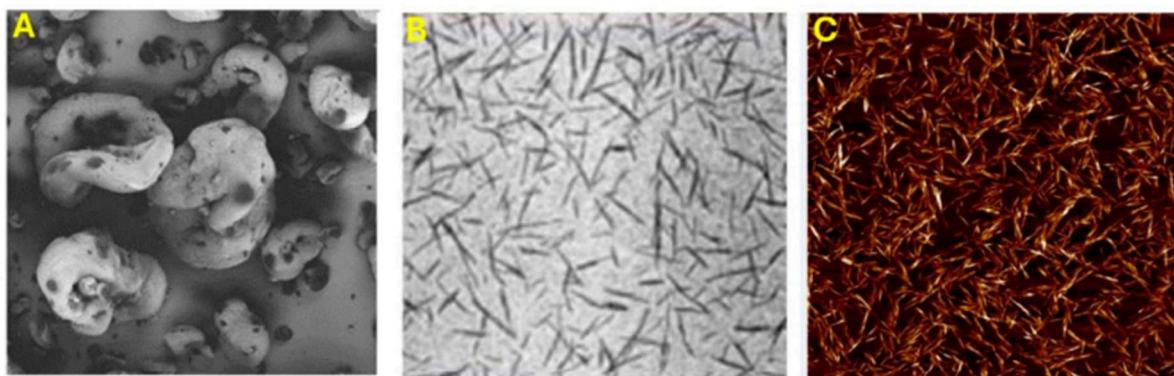


Figure 13. SEM, TEM, and AFM images of nanocrystalline cellulose (NCC). (A) SEM micrograph of spray-dried NCC powder; (B) TEM image showing rod-like cellulose nanocrystals; (C) AFM topographic image of individual nanocrystals, reproduced from reference [156]

Structurally, NCCs exhibit chemical anisotropy arising from variations in the density and orientation of surface hydroxyl groups. Regions with higher hydroxyl accessibility interact strongly with water, while less polar areas contribute a degree of hydrophobicity. This combination imparts mild amphiphilicity, enabling NCCs to adsorb at oil–water interfaces and function as solid stabilizers in Pickering emulsions [157].

Surface chemistry can be further tailored through post-functionalization reactions that expand the interfacial versatility of NCCs. Techniques such as silanization, esterification, amidation, and polymer grafting have been used to introduce chemical groups that modify surface wettability and dispersibility [148,158].

2.5.3. Applications in Emulsions

Recent research has advanced the use of nanocrystalline cellulose (NCC) as a renewable Pickering stabilizer for tailoring interfacial structure and stability in complex emulsions. Studies have shown that NCCs form rigid, non-desorbing layers at oil–water interfaces that provide exceptional resistance to droplet coalescence and Ostwald ripening [159].

Surface modification remains central to controlling emulsion type and curvature: hydrophilic sulfate-bearing NCCs preferentially stabilize oil-in-water emulsions, whereas hydrophobically modified or charge-neutral particles can invert the curvature to produce water-in-oil systems [157]. Introducing amphiphilic character by short-chain grafting or selective substitution of hydroxyl groups enhances the interfacial activity of NCCs and improves their ability to stabilize Pickering emulsions relative to untreated nanocrystals [160].

Combining NCCs with other colloids produces synergistic interfacial effects. Mixtures with biocolloids such as cellulose nanofibrils (CNFs) introduce depletion and network interactions that broaden stability ranges, with CNF–NCC systems showing robust droplet networks even at low CNF concentrations [161].

In addition, Lu *et al.* [162] showed that nanocellulose fiber flexibility critically governs emulsification behavior. Rigid CNCs form compact interfacial layers that stabilize droplets through repulsive and steric effects, whereas semi-flexible CNFs generate percolated networks that induce depletion-driven droplet clustering and gel-like rheology. In contrast, fully flexible bacterial cellulose lacks emulsifying capacity due to self-entanglement. These results demonstrate that tuning nanocellulose rigidity and aspect ratio enables precise control of emulsion microstructure and stability [162]

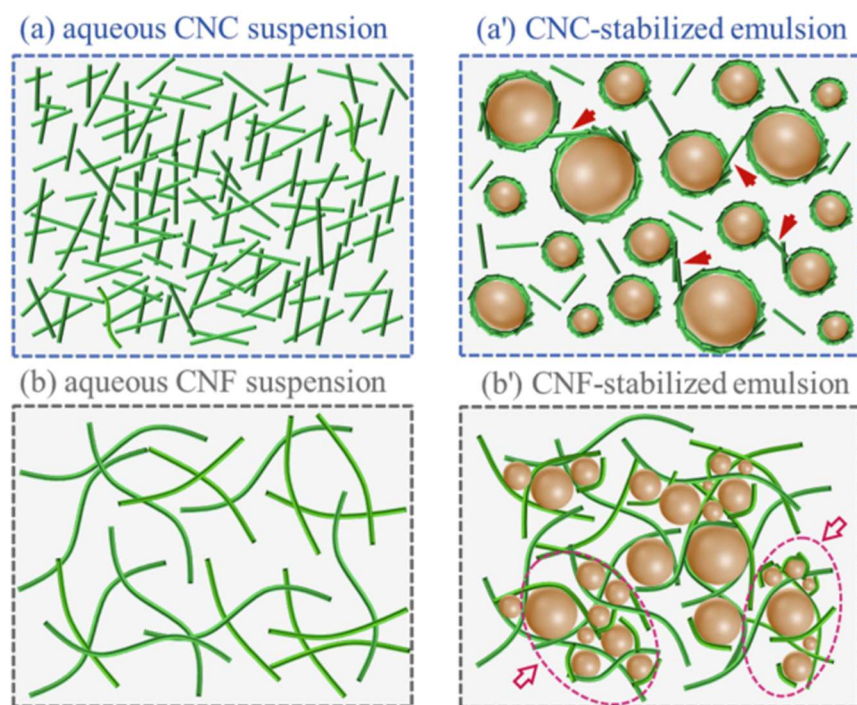


Figure 14. Schematic diagrams of percolated networks of (a, a') CNCs and (b, b') CNFs in aqueous suspensions and their corresponding Pickering emulsions reproduced from reference [162]

Tailoring the morphology of NCCs provides an additional means of controlling emulsion characteristics. Spherical NCCs, in particular, have been shown to enhance stability across a wide range of pH, ionic strength, and temperature, offering a means to influence interfacial packing and droplet deformation beyond what can be achieved by varying aspect ratio alone [163].

Recent studies have linked NCC concentration and morphology to the rheological properties of emulsions. Kinra and Pal [164] demonstrated that oil-in-water emulsions stabilized by NCC exhibit a progressive increase in viscosity and enhanced stability with rising NCC content, confirming its effectiveness as a robust interfacial stabilizer. [164].

NCCs have also been applied in more complex emulsion systems, including double emulsions stabilized by acylated nanocrystals for controlled release [165]. These systems demonstrate how controlled surface modification of NCCs can enable droplet organization and expand their utility in encapsulation and delivery applications.

These findings highlight the potential of NCCs to modulate emulsion architecture and stability across diverse systems.

2.6. Research Gaps Motivating This Thesis

Despite extensive studies on emulsion inversion and stability, several fundamental gaps remain in understanding how cellulose-based colloids influence catastrophic phase inversion (CPI) processes. Most previous work has focused on surfactant-stabilized systems, where inversion can be quantitatively described using thermodynamic frameworks such as the Hydrophilic–Lipophilic Deviation (HLD) and HLD–Net Average Curvature (HLD–NAC) models. These approaches capture formulation balance and interfacial curvature behavior but implicitly assume mobile, equilibrium-seeking interfaces, which do not adequately represent rigid, particle-based interfaces of Pickering emulsions.

In addition, most existing studies treat surfactant- and particle-stabilized emulsions as separate systems, with few direct comparisons performed under identical experimental conditions. This separation has limited the broader understanding of how different stabilization systems influence catastrophic phase inversion (CPI) behavior.

Furthermore, while NCC has been widely investigated as a renewable Pickering stabilizer, its role in governing inversion pathways and emulsion transition thresholds has received little attention.

Addressing these knowledge gaps is critical from a sustainability perspective. NCC offers a bio-based and non-toxic alternative to conventional stabilizers, yet its interfacial performance must be quantitatively benchmarked against classical surfactants to advance its broader adoption in industrial emulsification applications.

This work addresses these gaps through a direct comparison of catastrophic phase inversion in emulsions stabilized by nanocrystalline cellulose and by surfactants representing distinct chemical classes: anionic (SDS), cationic (OTAC), nonionic (Alfonic C12–14 alcohol ethoxylate), and zwitterionic (cetyl betaine, Amphosol). To the best of our knowledge, this study is the first to examine catastrophic inversion from water-in-oil to oil-in-water emulsions stabilized by nanocrystalline cellulose, offering new understanding of inversion behavior.

Chapter 3. Materials and Methods

3.1. Chemicals and Reagents

White mineral oil (Purity FG WO-15) was obtained from Petro-Canada (Mississauga, ON, Canada) and used as the continuous oil phase in all emulsion preparations. The oil has a kinematic viscosity of 15.0 cSt at 40 °C and 3.4 cSt at 100 °C.

Nanocrystalline cellulose (NCC) was supplied by CelluForce Inc. (Windsor, QC, Canada) under the trade name *CelluForce NCC® C100-NASD90*. The NCC powder consisted of rod-shaped particles with an average length of approximately 76 nm and an average width of 3.4 nm [166].

Four surfactants representing different headgroup charge classes were selected to enable systematic comparison of inversion behavior:

- **Anionic surfactant:** sodium dodecyl sulfate (SDS, ≥ 99 % purity, Fisher Scientific, Waltham, MA, USA; molecular weight = 288.38 g mol⁻¹).
- **Cationic surfactant:** octadecyltrimethylammonium chloride (OTAC, Molekula, Dallas, TX, USA; molecular weight = 348.05 g mol⁻¹).
- **Nonionic surfactant:** C12-14 alcohol ethoxylate with three ethylene oxide units (*Alfonic 1412-3*, Sasol Chemicals, Sandton, South Africa; average molecular weight 336 g mol⁻¹).
- **Zwitterionic surfactant:** cetyl betaine (*Amphosol*, Stepan Company, Northfield, IL, USA), containing both quaternary ammonium and carboxylate groups; molecular weight = 327.61 g mol⁻¹.

A detailed description of the molecular structures is provided in Section 2.2 (Surfactant Chemistry and Stabilization Mechanisms). All surfactants were used as received without further purification. Deionized water was used in all preparations, and sodium chloride (≥ 99.5 % purity, Fisher Scientific) was added to the aqueous phase when necessary to adjust conductivity for phase inversion measurements.

3.2. Preparation of NCC Dispersion

Nanocrystalline cellulose (NCC) was dispersed in deionized water using a variable-speed rotor–stator homogenizer (Model 1-L, Gifford–Wood, Hudson, NY, USA) equipped with an open slotted stator head designed for high-shear mixing. A batch volume of 2 L was prepared at room temperature, and NCC was added gradually to achieve concentrations ranging from 0 to 1.5 wt% relative to the aqueous phase.

The dispersion was first premixed at 2000 rpm for 5 min to ensure proper wetting and breakup of agglomerates, followed by high-shear homogenization at 5000 rpm until a uniform and stable dispersion was obtained. Dispersion quality was confirmed by visual inspection for the absence of visible aggregates or flocs.

After homogenization, sodium chloride (99.5% purity, Fisher Scientific) was added to the aqueous phase to impart ionic conductivity, enabling detection of the phase inversion point during conductivity measurements. The resulting NCC dispersions were used immediately as the aqueous phase for emulsion preparation.

3.3. Phase Inversion Experiments

Phase inversion experiments were conducted to determine the effect of nanocrystalline cellulose (NCC) and surfactant type on the transition from water-in-oil (W/O) to oil-in-water (O/W) emulsions. The experiments were performed using the same rotor–stator homogenizer (Model 1-L, Gifford–Wood, Hudson, NY, USA) operated under controlled mixing conditions.

The aqueous phase, containing either NCC or a surfactant of fixed concentration, was gradually added to a known volume of oil (Purity FG WO-15, 600 mL) under continuous agitation. Additions were performed at 2–3 min intervals to ensure uniform mixing between each increment. The aqueous phase volume was increased stepwise up to approximately 1500 mL, corresponding to a maximum final aqueous volume fraction of about 0.7.

Conductivity measurements were recorded after each addition, including beyond the inversion point, to capture the complete conductivity profile during emulsification. The phase inversion point was identified by a sharp increase in electrical conductivity, corresponding to the transition from a W/O to an O/W emulsion.

The homogenizer speed was initially maintained at 4000 rpm and gradually increased to 5000 rpm to compensate for viscosity buildup as the emulsion approached inversion. Stator geometry, mixing time, and addition protocol were kept constant across all runs to maintain comparable shear energy input and minimize variation in droplet size distribution. While droplet size was not measured directly, this procedure ensured uniform shearing conditions throughout the emulsification process and consistent determination of the phase inversion point.

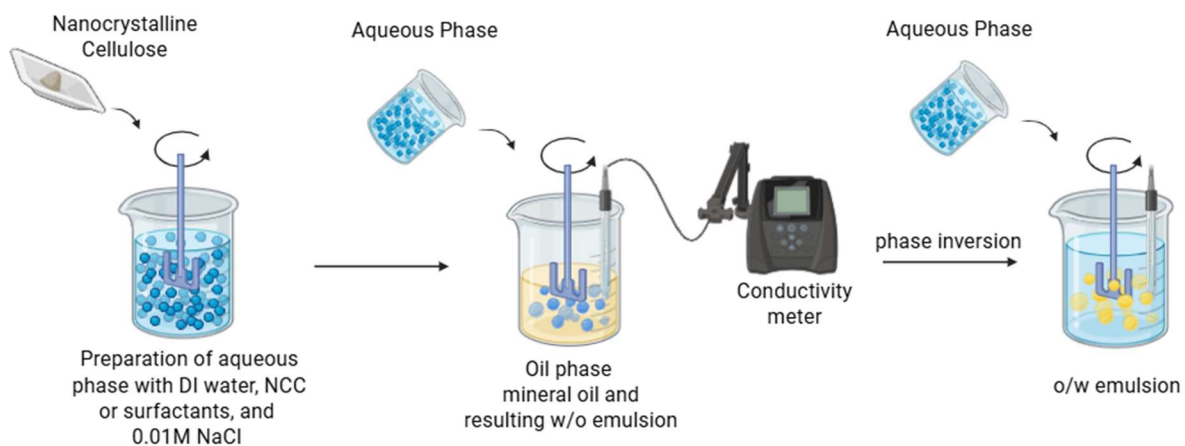


Figure 15. Schematic of phase inversion experiment



Figure 16. Laboratory set up for the phase inversion experiments

3.4. Stability Experiments

Emulsion stability was evaluated by monitoring coalescence behavior over time. Water-in-oil (W/O) emulsions containing a fixed aqueous phase volume were prepared by homogenizing oil with aqueous solutions of either NCC or surfactants at high shear for 5 min. Unless otherwise noted, the aqueous phase volume was set to 20 vol%, except in the case of SDS, where 10 vol% was used due to its higher interfacial activity and consequently quicker phase inversion.

Immediately after emulsification, samples were transferred into 500 mL graduated cylinders and stored at ambient temperature without agitation. The extent of phase separation was measured periodically by recording the volume of the coalesced aqueous layer at the bottom of each cylinder.

Emulsion stability was quantified as the ratio of the separated aqueous phase volume to the total volume.

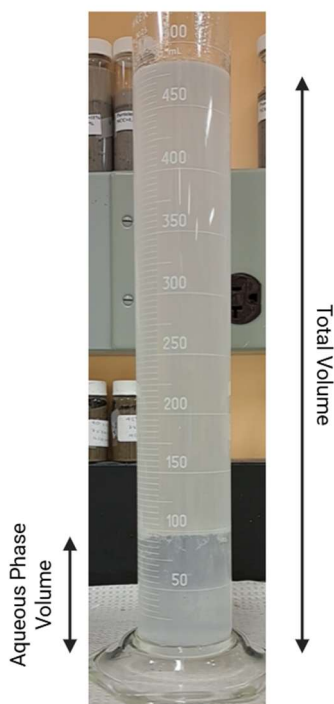


Figure 17. Coalescence stability testing set-up; emulsion stability is determined based on the aqueous phase (bottom layer) volume relative to the total volume measured as a function of time.

3.5. Surface and Interfacial Tension Measurements by Pendant Drop Tensiometry

Interfacial and surface tension measurements were performed using the pendant drop method with a smartphone-based tensiometer (Droplet Lab, Markham, ON, Canada). Drop shape analysis was conducted by fitting the droplet profile to the Young–Laplace equation using axisymmetric drop shape analysis (ADSA) [167].

A pendant droplet of the aqueous phase (typically 10–20 μL), containing nanocrystalline cellulose (NCC) or one of the surfactants, was formed at the tip of a stainless-steel needle (1.8 mm diameter) connected to a 500 μL Hamilton® gastight syringe (Model 1750 TPLT, Hamilton, Reno, NV, USA). The droplet was dispensed using a screw-driven plunger to allow precise control of flow rate and drop formation. A uniform backlight provided by an LED source ensured consistent optical contrast during image capture.

For interfacial tension (IFT) measurements, droplets were dispensed into a sealed quartz cuvette containing the oil phase (Purity FG WO-15). For surface tension measurements, the droplet was suspended in air under identical optical and geometric conditions. Each sample was measured ten times to ensure reproducibility, and the mean value was reported. All experiments were conducted at ambient temperature (22 ± 1 °C).

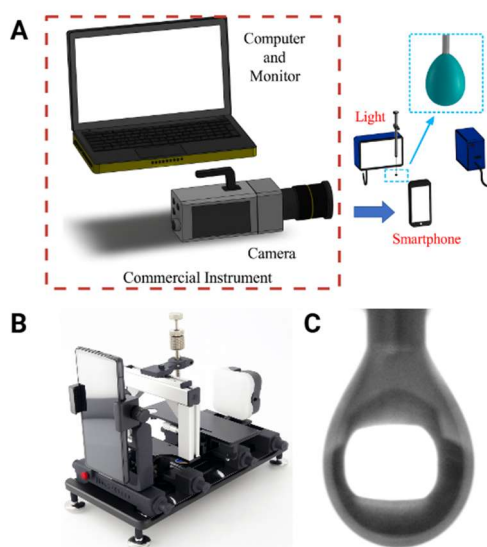


Figure 18. a) Schematic of portable tensiometer compared with commercial instrument reproduced from reference [167], b) DropletLab's tensiometer model, c) Image of NCC solution capture by the tensiometer to determine the SFT

Chapter 4. Results

4.1. Phase Inversion Behavior of NCC-Stabilized Emulsions

The catastrophic phase inversion behavior of water-in-oil (W/O) emulsions stabilized by nanocrystalline cellulose (NCC) was investigated by progressively increasing the aqueous phase fraction under controlled shear conditions. In these experiments, the concentration of NCC in the aqueous phase was varied from 0 to 1.5 wt%. Conductivity measurements were used to track the progression of inversion and identify the emulsion inversion point (EIP), defined as the critical aqueous phase volume fraction where a sharp increase in conductivity indicated the transition from a non-conductive W/O system to a conductive oil-in-water (O/W) emulsion.

In the absence of NCC, the EIP was observed at an aqueous volume fraction of approximately 0.25, characteristic of low resistance to inversion and limited interfacial stability. Upon addition of NCC, the system exhibited a pronounced shift in inversion behavior, with the EIP increasing with particle concentration. At 0.1 wt% NCC, the inversion point increased to 0.32, and further increases to 0.2, 0.3, and 0.5 wt% shifted it to 0.40, 0.44, and 0.47, respectively. Beyond 0.5 wt%, the increase became progressively smaller, reaching 0.55 at 1.5 wt%. The conductivity data are shown in Figure 19. wt%, the increase became progressively smaller, reaching 0.55 at 1.5 wt%. The conductivity data are shown in Figure 19.

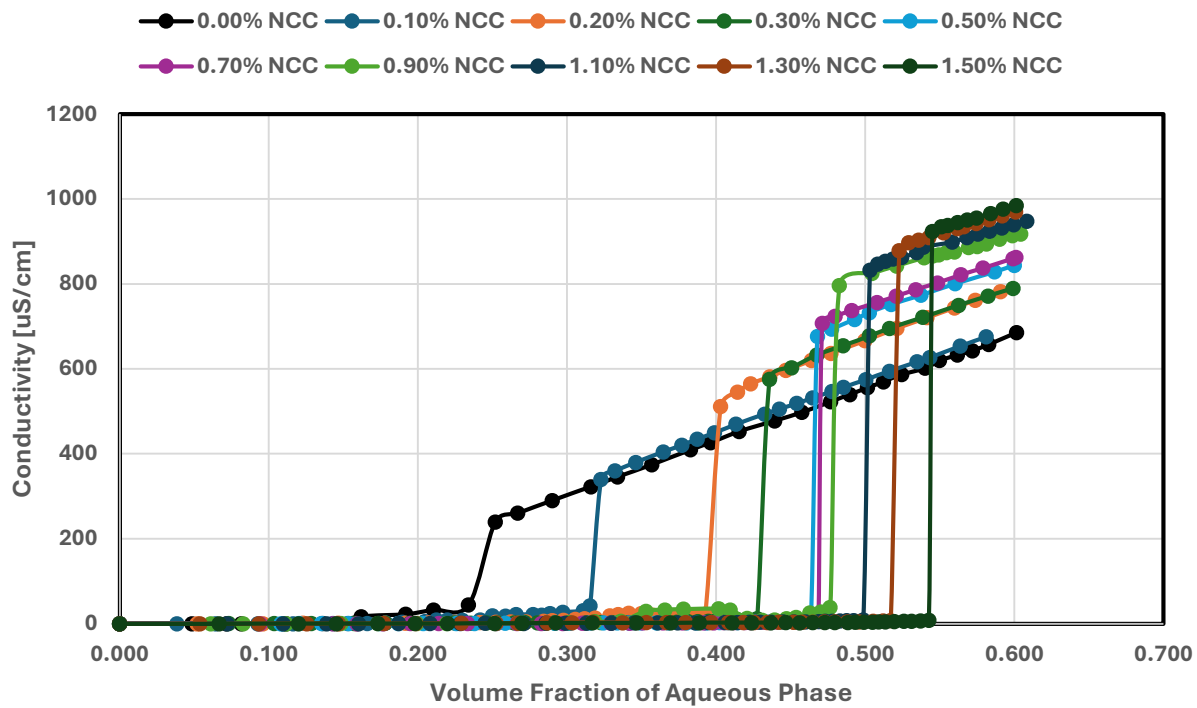


Figure 19. Phase inversion of W/O to O/W emulsion at different NCC concentrations ranging from 0 to 1.5 wt%

The corresponding variation of the emulsion inversion point with NCC concentration is plotted in Figure 20. As shown, the EIP increases nonlinearly with NCC loading, displaying a steep initial rise at low concentrations followed by a gradual plateau beyond 0.5 wt%.

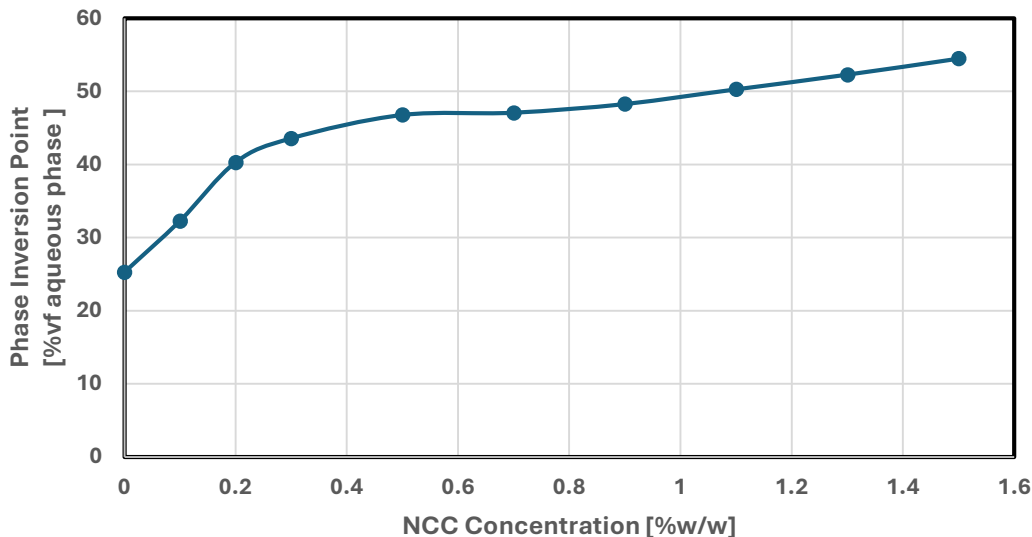


Figure 20. Phase inversion point as a function of NCC concentration (%wt).

This nonlinear dependence suggests two stabilization regimes. At low particle loadings, additional NCC markedly enhances interfacial coverage and suppresses coalescence by forming a continuous barrier around dispersed droplets. As concentration increases, the interface becomes progressively saturated, and further addition of NCC contributes less to stabilization because of particle crowding and limited available surface area. The slope reduction in Figure 20 therefore indicates a transition from a coverage-controlled regime to a jammed-interface regime, where the resistance to inversion is governed by interfacial elasticity and rigidity, rather than by additional surface coverage.

The observed trend reflects the progressive buildup of particle packing of the interfacial network. NCCs forms rigid, elastic film that resists deformation and droplet coalescence, typical of pickering emulsion networks, due to its high aspect ratio and surface charge. The adsorption is relatively irreversible and generate strong steric hindrance and interfacial viscoelasticity, effectively suppressing the rearrangements of droplets required for phase transition. Consequently,

a significantly larger phase fraction is needed to overcome the mechanical strength of the jammed network and trigger inversion.

The resistance to inversion observed here mirrors that reported in other Pickering systems. For instance, Bains and Pal [121] found that starch nanoparticles elevated the inversion point of W/O emulsions beyond 0.5 aqueous fraction [121], while Binks and Lumsdon [69] observed that hydrophobic silica nanoparticles delayed inversion until water fractions approached 0.7 [69]. The similarity of these results confirms that the mechanism is dominated by interfacial jamming and mechanical rigidity rather than by interfacial-tension effects.

Collectively, these findings establish that NCC acts as a highly effective Pickering stabilizer capable of delaying catastrophic phase inversion by reinforcing the interfacial film and suppressing droplet rearrangement under shear. Such behavior sharply contrasts with the trends observed for surfactant-stabilized emulsions discussed in Section 4.2, where increasing surfactant concentration promotes earlier inversion through interfacial saturation and dynamic redistribution.

4.2. Phase Inversion Behavior of Surfactant-Stabilized Emulsions

In contrast to the gradual and concentration-dependent delay observed in NCC-stabilized emulsions, systems stabilized by molecular surfactants exhibited markedly different catastrophic phase inversion behavior.

Emulsions were prepared using four surfactants representing distinct headgroup chemistries— anionic (sodium dodecyl sulfate, SDS), cationic (octadecyltrimethylammonium chloride, OTAC), nonionic (C12–14 alcohol ethoxylate, Alfonic), and zwitterionic (cetyl betaine, Amphosol). Each surfactant was dissolved in the aqueous phase at concentrations ranging from 0.1 to 1 wt %, and the aqueous phase was incrementally introduced into the mineral-oil continuous phase under identical mixing conditions used for NCC experiments. Conductivity and initiate curvature reversal was measured after each addition to determine the EIP.

The electrical conductivity profiles for each surfactant are shown in Figures 21–24, while the corresponding inversion points as a function of surfactant concentration are summarized in Figure 25. At low surfactant concentrations, all systems exhibited a delayed inversion relative to the surfactant-free control, with the EIP shifting to higher aqueous volume fractions. This trend is somewhat counter-intuitive given the hydrophilic nature of SDS, OTAC, C12-C14 alcohol ethoxylate, and cetyl betaine which typically favor oil-in-water (O/W) emulsions based on Bancroft's rule.

This behavior indicates that at low concentrations, surfactant molecules provide limited yet sufficient interfacial coverage to stabilize the existing W/O structure, suppressing coalescence and delaying phase inversion despite their inherent affinity for the aqueous phase. The delayed transition suggests that a critical surfactant concentration is required before the molecules can effectively initiate curvature reversal and promote oil-in-water emulsification. At concentrations below this threshold, insufficient interfacial coverage likely yields only transient kinetic stabilization of the existing water-in-oil structure delaying inversion until droplet crowding and hydrodynamic stresses overcome the metastable state.

As surfactant concentration increased, a distinct reversal in trend was observed. Beyond a concentration-specific threshold, the inversion point shifted to progressively lower aqueous phase fractions, signifying an earlier onset of inversion. With greater surfactant availability, adsorption

at the oil–water interface becomes rapid and extensive, lowering interfacial tension and facilitating the curvature reversal required for O/W formation. Once the interface becomes saturated, excess surfactant molecules partition into the bulk or form micelles that act as curvature modifiers, further promoting inversion at lower internal-phase fractions.

This transition is consistent across surfactant classes, though the magnitude of the shift varies with headgroup

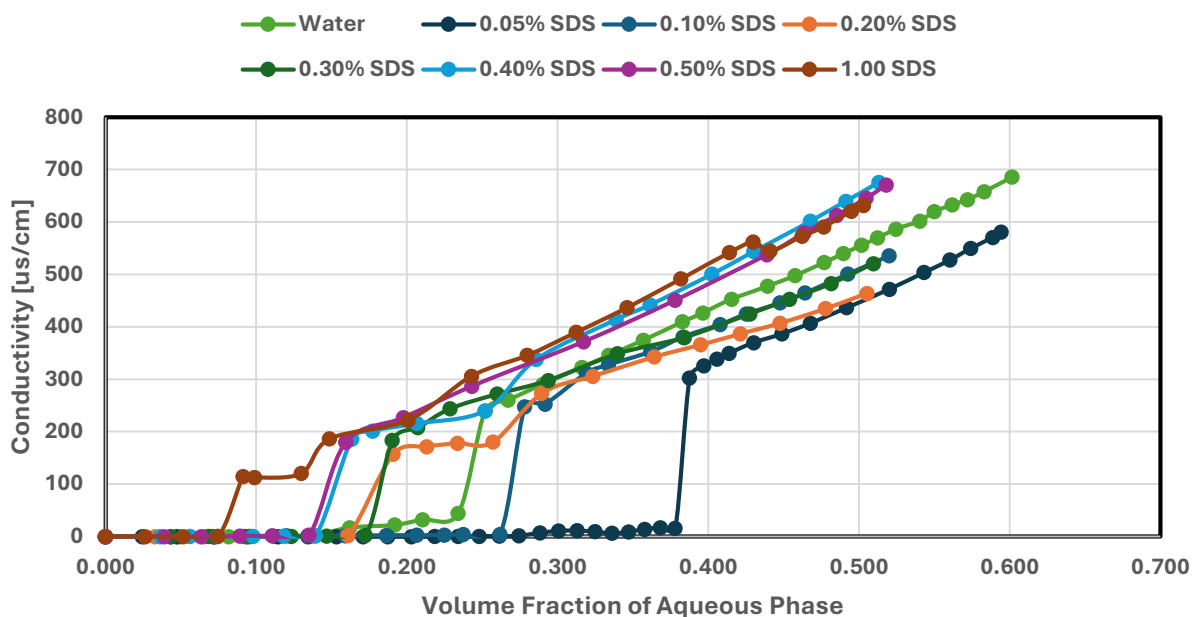


Figure 21. Phase inversion of W/O to O/W emulsion at different SDS concentrations

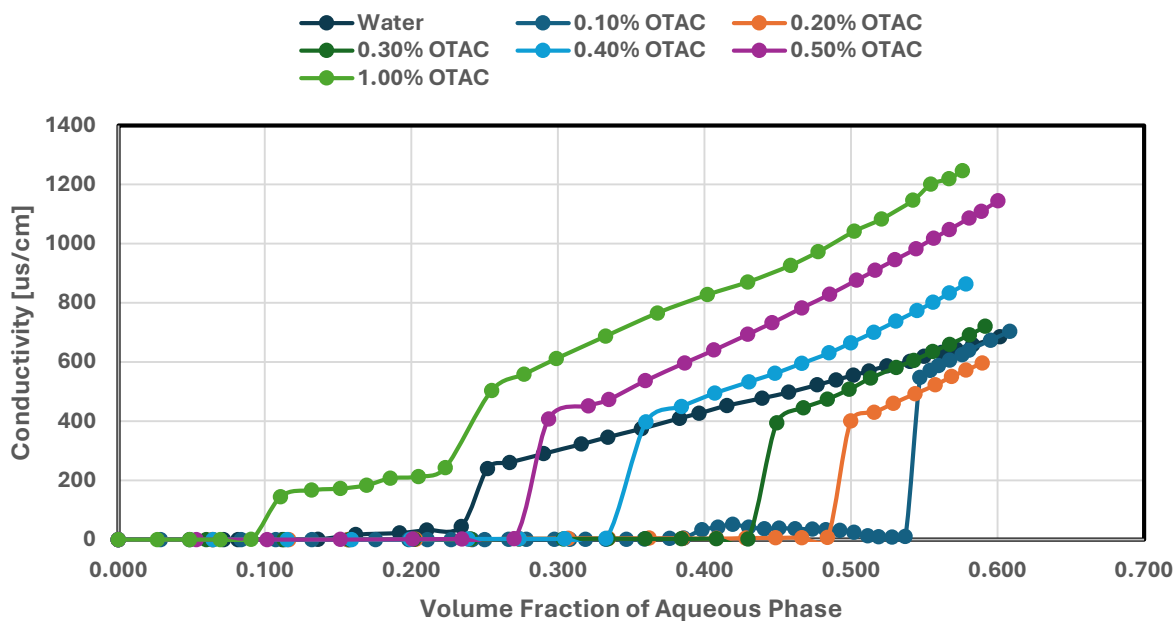


Figure 22. Phase inversion of W/O to O/W emulsion at different OTAC concentrations

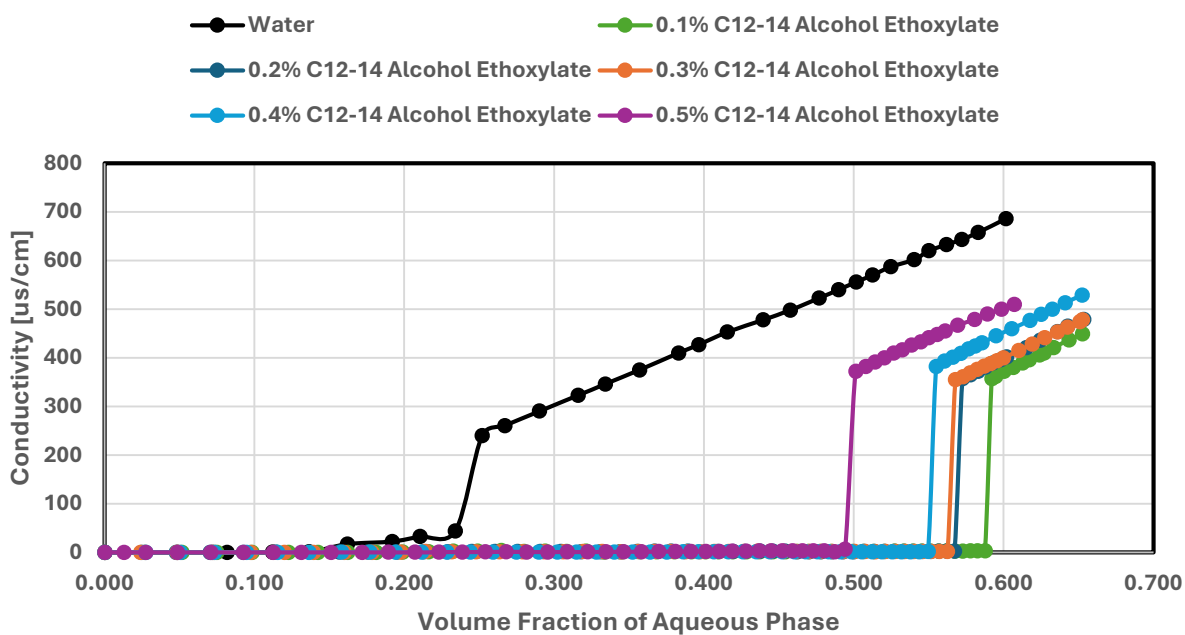


Figure 23. Phase inversion of W/O to O/W emulsion at different C12-C14 alcohol ethoxylate concentrations

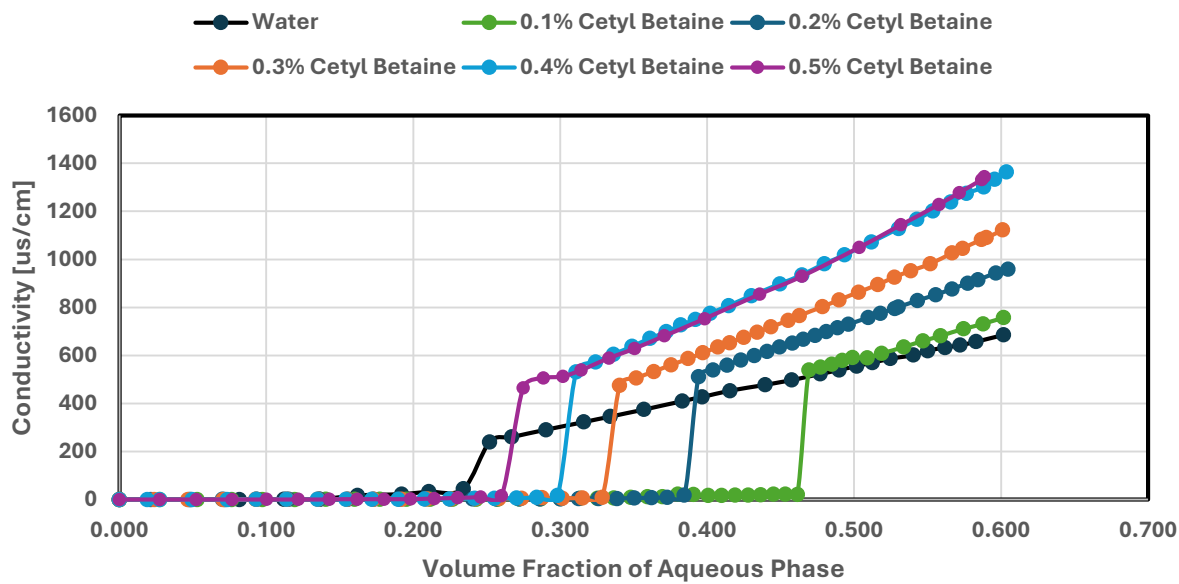


Figure 24. Phase inversion of W/O to O/W emulsion at different cetyl betaine concentrations

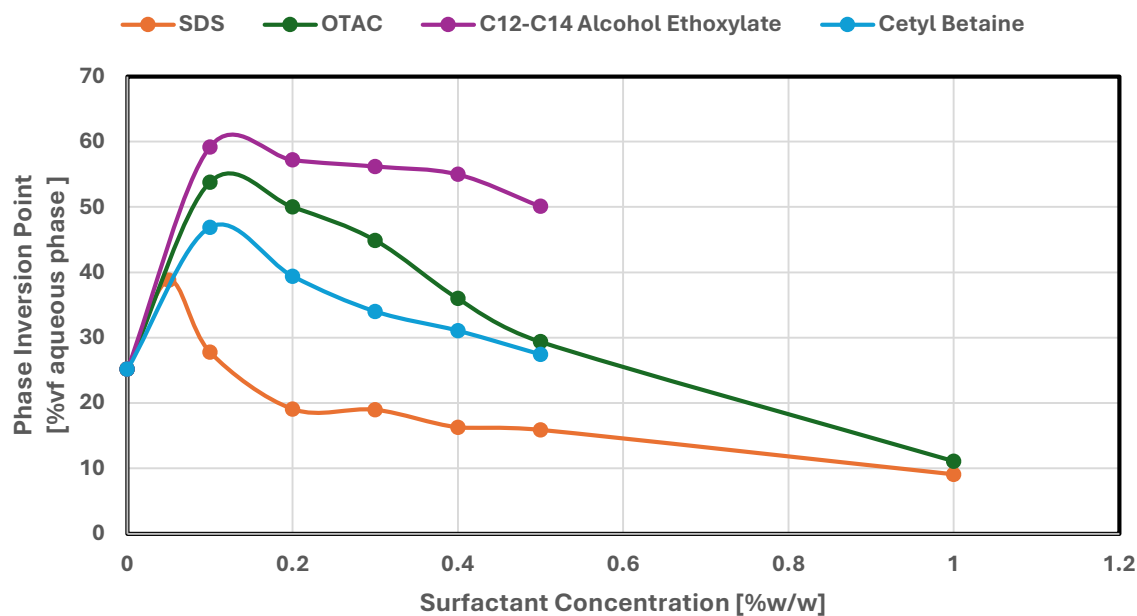


Figure 25. Comparison of phase inversion of W/O to O/W emulsion for different surfactants

The non-monotonic dependence of EIP on surfactant concentration therefore reflects competing kinetic and thermodynamic effects. At low concentrations, the system is dominated by kinetic stabilization where insufficient surfactant coverage restricts droplet coalescence and temporarily

maintains a metastable W/O configuration. At higher concentrations, the behavior becomes thermodynamically driven, as the reduced interfacial tension and excess surfactant favor the formation of a continuous aqueous phase.

At higher concentrations, the ionic surfactants, most notably SDS and then OTAC, produce the strongest shift to early inversion, with the EIP dropping to the lowest values. Cetyl betaine shows an intermediate response. In contrast, the nonionic alcohol ethoxylate maintains elevated EIP over the tested range, indicating a more persistent delay to inversion. This trend highlights fundamental differences in how ionic and nonionic surfactants organize and evolve at the oil–water interface, as discussed further in Section 5.1.

In summary, the results demonstrate that surfactant-stabilized systems display a distinct two-regime response: (i) delayed inversion at sub-critical concentrations due to limited coverage and kinetic stabilization, and (ii) accelerated inversion at supra-critical concentrations driven by interfacial saturation and curvature reversal.

Notably, these findings emphasize that the contrasting inversion behavior between particle- and surfactant-stabilized emulsions originates from fundamentally different modes of interfacial stabilization.

4.3. Surface Tension and Interfacial Tension

Surface and interfacial tension measurements provide direct insight into the interfacial activity and adsorption behavior of NCC and the four molecular surfactants. These properties govern how efficiently stabilizers populate the oil–water boundary and thereby influence the catastrophic inversion trends discussed previously.

4.3.1. Surface Tension

The equilibrium surface tension of aqueous phases containing NCC or surfactants is shown in Figure 25. The surface tension results reveal a clear distinction between NCC- and surfactant-based stabilizers.

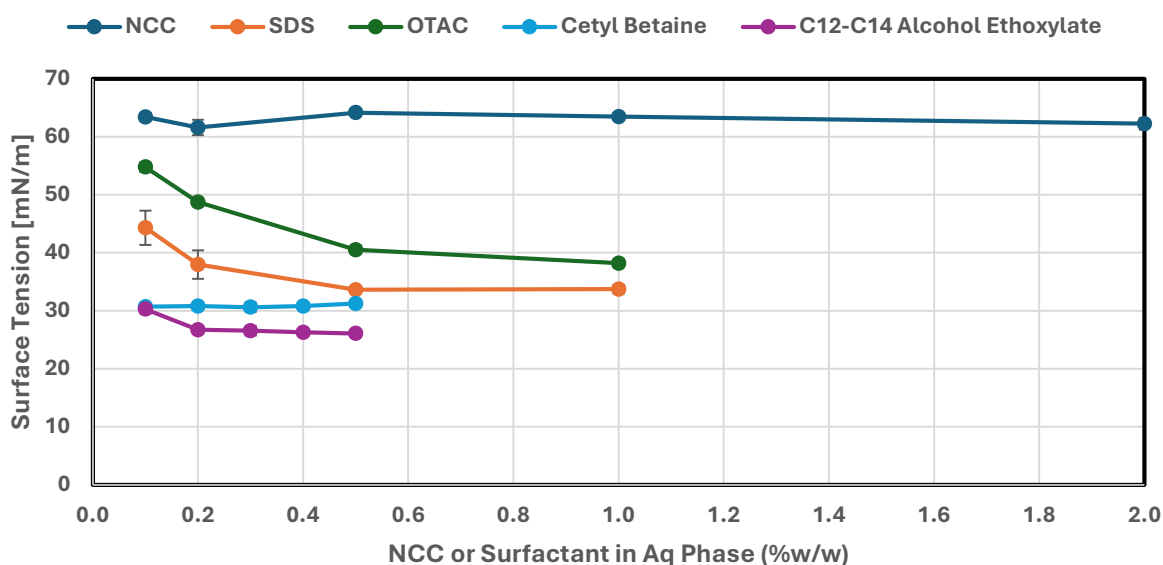


Figure 26. Surface tension of NCC and surfactants (SDS, OTAC, cetyl betaine, C12-C14 alcohol ethoxylate) as a function of NCC and surfactant concentration

Aqueous dispersions of NCC maintained high surface tension values (≈ 63 mN/m) across the entire concentration range, decreasing only slightly from 63.4 mN/m at 0.1 wt% to 62.3 mN/m at 2.0 wt%, with minimal variation between intermediate concentrations. The weak surface activity reflects the rigid, colloidal nature of NCC particles and their limited molecular mobility at the air–water interface. Stabilization in NCC systems therefore arises primarily from steric hindrance and

the formation of mechanically robust interfacial layers at the oil-water interface rather than from dynamic tension reduction.

In contrast, all four surfactants exhibited pronounced reductions in surface tension with increasing concentration. SDS exhibited a sharp decrease from 44.23 mN/m at 0.1 wt% to 33.74 mN/m at 1.0 wt%, while OTAC followed a similar trend, decreasing from 54.80 mN/m to 38.22 mN/m over the same range. In comparison, cetyl betaine and C12-C14 alcohol ethoxylate achieved more pronounced overall reductions, reaching 31.25 mN/m and 26.11 mN/m, respectively, at 0.5 wt%; however, the rate of decrease with concentration was less steep than that observed for the ionic surfactants, indicating a more gradual adsorption process and slower approach to equilibrium surface coverage. These results demonstrate that all surfactants actively reduce interfacial free energy, though the extent depends on their molecular structure and headgroup chemistry.

4.3.2. Interfacial Tension

The interfacial tension (IFT) data further highlights the contrasting stabilization mechanisms of particles and surfactants, as shown in Figure 27.

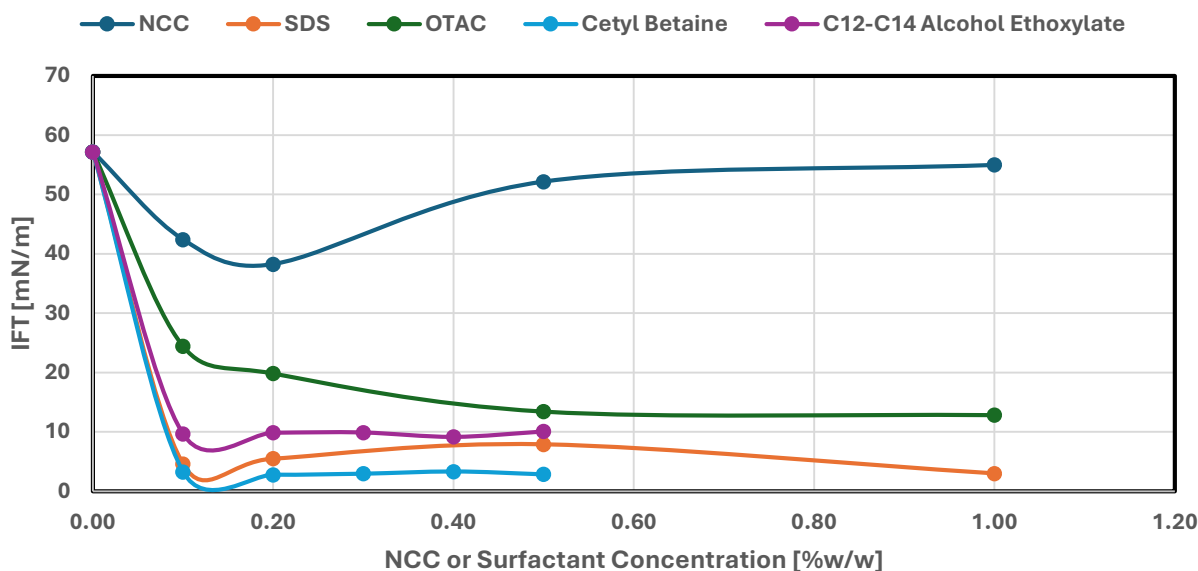


Figure 27. Interfacial tension of oil and aqueous phase consisting of NCC and surfactants at various concentrations.

NCC exhibited a limited ability to reduce oil–water interfacial tension across the tested concentration range. An initial decrease in IFT was observed at low concentrations (0.1–0.2 wt%),

reaching a minimum of 38.3 mN/m. This reduction is likely due to partial adsorption of NCC particles at the interface, where their amphiphilic surface character and rod-like geometry enable them to anchor and displace water–oil contact, thereby lowering interfacial energy. However, beyond this concentration range, IFT begins to plateau or slightly increase, indicating more complex interfacial behavior. This may be attributed to particle crowding, bulk aggregation, or kinetic limitations that restrict further adsorption. Alternatively, interfacial rearrangement or multilayer formation could disrupt the uniformity and stability of the interfacial film. The non-monotonic

IFT response observed for NCC suggests that while partial adsorption lowers interfacial energy initially, additional particles contribute minimally to further stabilization once the interface becomes saturated.

In contrast, the surfactant systems showed dramatic reductions in interfacial tension. SDS reduced IFT from 57.2 mN/m in the absence of surfactant to 4.61 mN/m at 0.1 wt % and reached 3.02 mN/m at 1 wt % reflecting its strong interfacial activity and rapid adsorption kinetics which are well-established. Cetyl betaine displayed similarly strong performance, lowering IFT to 2.81 mN/m at 0.2 wt % and maintaining values below 3.4 mN/m across the full range. C12-14 alcohol ethoxylate exhibited more moderate reductions, stabilizing near 10 mN/m at all concentrations tested, while OTAC showed a slower but steady decrease, falling from 24.46 mN/m at 0.1 wt % to 12.86 mN/m at 1 wt %. As illustrated in Figure 27, all surfactants produced a steep initial decline in IFT at low concentrations, reflecting efficient adsorption at the oil–water interface where even small amounts of surfactant significantly reduce interfacial energy.

This rapid early drop coincides with the concentration range that delays catastrophic inversion (Section 4.2), suggesting that rapid surfactant adsorption enhances interfacial stability by suppressing coalescence and extending the persistence of the W/O structure. At higher surfactant concentrations, a threshold is reached where the interface becomes saturated and further decreases in IFT are minimal. Beyond this point, excess surfactant accumulates in the bulk or micellar phase rather than reinforcing the interface. This behavior suggests that, under high shear, the system becomes more prone to catastrophic inversion because of surplus surfactants within the continuous phase.

4.3.3. Comparative Implications

The surface- and interfacial-tension measurements clearly distinguish two fundamentally different modes of emulsion stabilization. The sharp, concentration-dependent reductions in SFT and IFT observed for surfactant systems confirm that their stability originates from dynamic modulation of interfacial energy. Rapid adsorption and desorption of surfactant molecules enable continuous adjustment of curvature and promote phase transitions once interfacial saturation is reached. In contrast, the consistently high SFT and limited IFT reduction measured for NCC demonstrate that its stabilization arises not from energetic minimization but from the formation of mechanically persistent interfacial films which likely contribute to the delayed phase inversion. In summary, the SFT and IFT data show that surfactant-stabilized emulsions invert readily upon reaching minimum interfacial tension, whereas NCC-stabilized systems remain stable due to the inherent rigidity of their interfacial particle networks.

4.4. Coalescence Stability

4.4.1. Nanocrystalline Cellulose

The coalescence stability experiments highlighted clear differences in how nanocrystalline cellulose and molecular surfactants maintain emulsion stability over time. The progression of phase separation over time revealed how each system resisted droplet coalescence as illustrated in Figure 28.

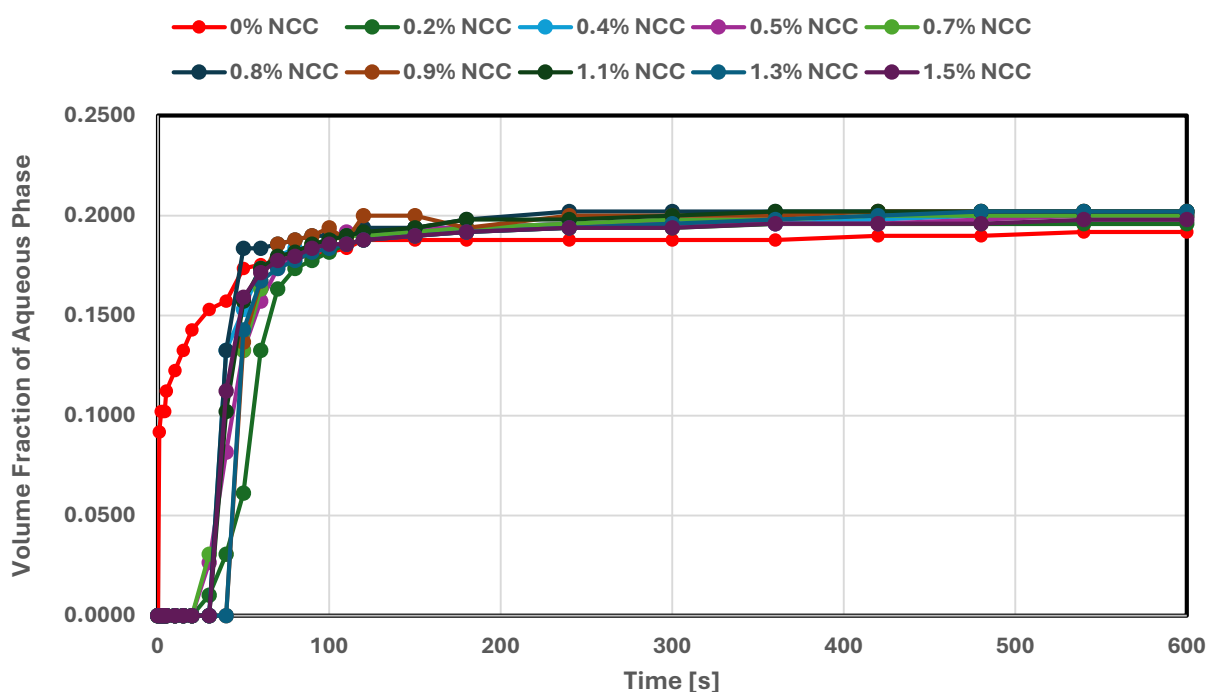


Figure 28. Coalescence stability of W/O emulsion of oil and NCC solution with varying NCC concentration

At 0.2 wt%, the lowest NCC concentration tested, the emulsion exhibited delayed coalescence compared with the 0 wt% control, indicating that low particle loading is sufficient to provide measurable resistance to droplet merging. At concentrations above 0.2 wt%, the aqueous phase volume fraction increased consistently across all samples, suggesting convergence in coalescence rate and implying that the interface became saturated with particles such that additional NCC contributed minimally to further stability. These results confirm that NCC imparts steric stabilization that is primarily concentration-dependent up to the percolation threshold, beyond which interfacial crowding limits further gains in coalescence resistance.

Surfactant-stabilized systems displayed more complex, surfactant-specific behaviors, as described below.

4.4.2. Anionic Surfactant - SDS

For SDS-stabilized emulsions, coalescence stability exhibited a nonlinear trend with increasing surfactant concentration (Figure 29). Since SDS-stabilized systems were known to undergo catastrophic phase inversion before reaching 20 vol% aqueous phase (as determined in Section 4.1), aqueous volume fraction of 10 vol% was used for the coalescence tests. At 0.0% SDS, the aqueous phase began separating early and stabilized near a volume fraction of approximately 0.09, indicating poor emulsion stability. At all SDS concentrations, phase separation of the aqueous layer was delayed relative to the control. The inclusion of a low SDS concentration (0.05 wt%) captured the early stabilization regime observed prior to inversion, as discussed in Section 4.1, showing that even trace surfactant addition substantially delayed coalescence relative to the control. At this level, the emulsion exhibited the slowest coalescence rate and the smallest final separated volume fraction. Above 0.05 wt%, the coalescence of the aqueous layer proceeded more rapidly; however, the total separated volume fraction in these samples exceeded the nominal 10% loading. This overestimation is attributed to the formation of a stable, cream-like emulsion layer that visually overlapped with the aqueous phase, making it difficult to distinguish a clear phase boundary during measurement. Despite this, the overall trend remained evident that SDS improved the emulsion stability at all concentrations relative to the control.

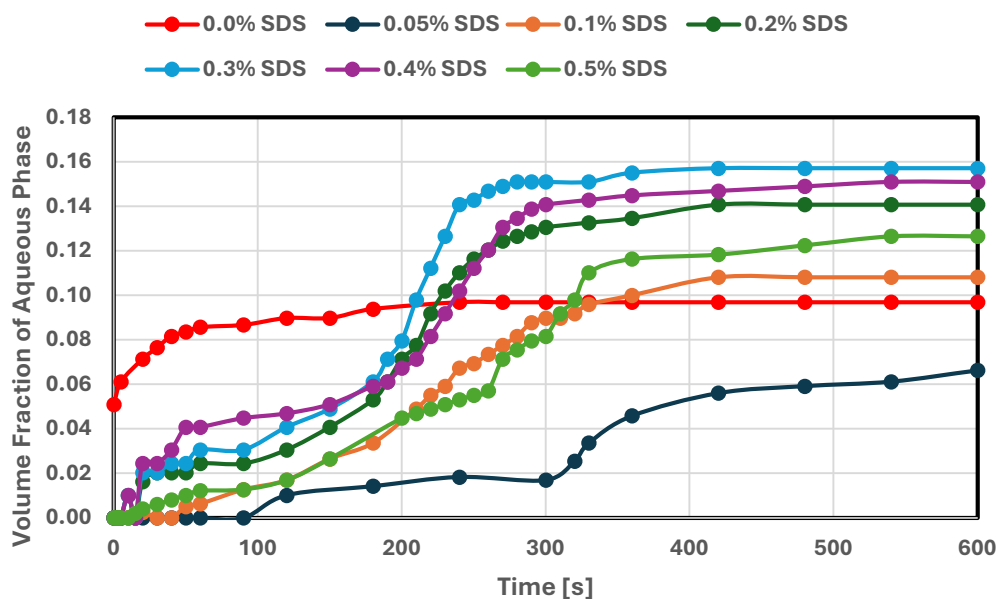


Figure 29. Coalescence stability of W/O emulsion of oil and SDS solution with varying SDS concentration.

4.4.3. Cationic Surfactant - OTAC

OTAC-stabilized emulsions showed consistent suppression of coalescence across all concentrations tested (Figure 30). Even at the lowest concentration (0.1 wt%), OTAC effectively delayed phase separation compared with the control, indicating that minimal cationic surfactant loading was sufficient to impart strong interfacial stability. The 0.1 and 0.2 wt% formulations exhibited nearly identical stability profiles, suggesting early saturation of the interface or comparable packing efficiency at these concentrations. At 0.3 and 0.4 wt%, a further delay in aqueous phase separation was observed, with both concentrations showing similar performance, likely reflecting a plateau in stabilization behavior. At 0.5 wt%, the measured aqueous phase volume exceeded the nominal 20% loading, attributed to the formation of a dense, cream-like emulsion layer that overlapped visually with the separated phase, which is an effect also observed in the SDS system. Overall, OTAC effectively delayed coalescence across all concentrations tested, maintaining stable emulsions even at low surfactant levels due to its strong interfacial adsorption and compact molecular packing.

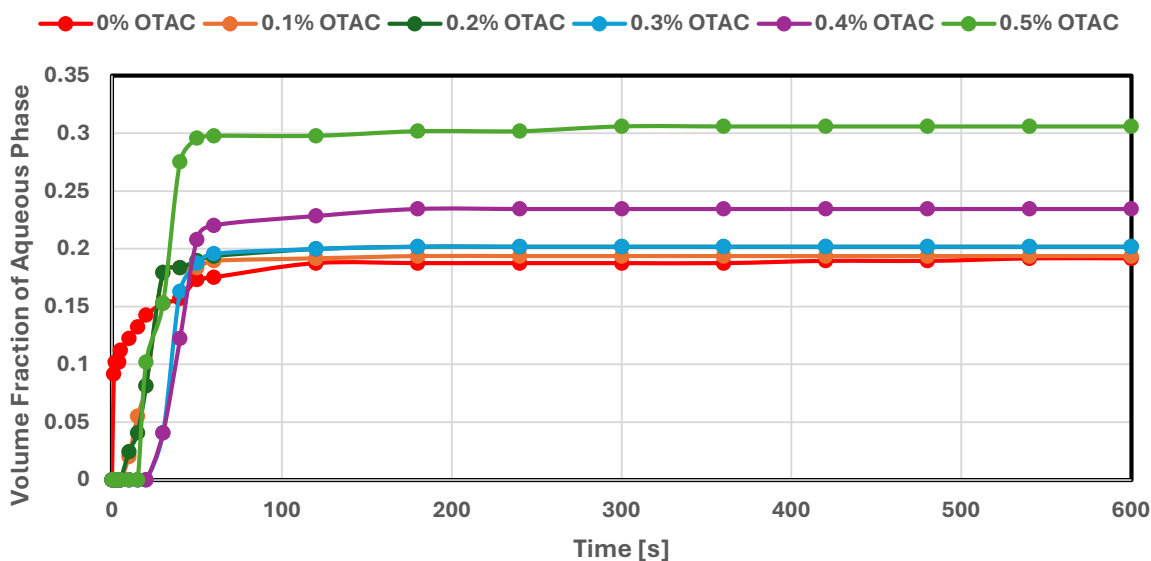


Figure 30. Coalescence stability of W/O emulsion of oil and OTAC solution with varying OTAC concentration

4.4.4. Non-ionic Surfactant – Alcohol Ethoxylate

Alcohol ethoxylate-stabilized emulsions demonstrated consistent resistance to coalescence across the concentration range tested (Figure 31). At low concentrations (0.1–0.2 wt%), emulsions exhibited delayed phase separation compared with the 0 wt% control, indicating that relatively small amounts of the nonionic surfactant were sufficient to form a stable interfacial layer. As the concentration increased to 0.3–0.5 wt%, only marginal improvement in coalescence stability was observed, suggesting that the interface had reached saturation within this range. Beyond this point, additional surfactants likely remained in the bulk phase rather than contributing to further interfacial stabilization. The stable behavior across all concentrations reflects the inherent effectiveness of Alfonic in maintaining cohesive interfacial films that provide lasting resistance to droplet coalescence and ensure extended emulsion stability.

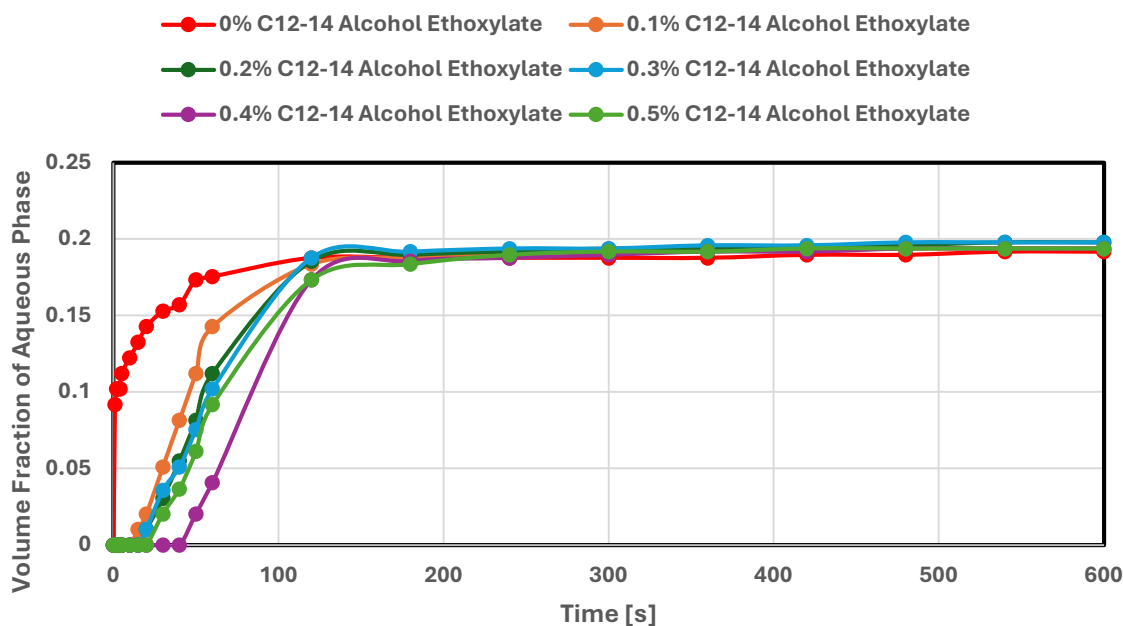


Figure 31. Coalescence stability of W/O emulsion of oil and C12-C14 alcohol ethoxylate solution with varying C12-14 alcohol ethoxylate concentration

4.4.5. Zwitterionic Surfactant – Cetyl Betaine

Cetyl Betaine-stabilized emulsions exhibited a non-monotonic trend in coalescence stability across the tested concentration range (Figure 31). Relative to the control, emulsions containing 0.1–0.2 wt% Amphosol showed a marked improvement in stability, displaying the slowest coalescence rate. As the concentration increased beyond 0.2 wt%, stability gradually declined, with 0.3–0.5 wt% samples separating more rapidly, though still performing better than the surfactant-free system. The zwitterionic nature of cetyl betaine likely contributes to its strong stabilization at low concentrations by enabling balanced electrostatic interactions that minimize repulsion and favor dense adsorption. At higher loadings, however, oversaturation and altered headgroup alignment may reduce these stabilizing effects. Overall, cetyl betaine provided effective stabilization across the entire concentration range, with optimal coalescence resistance achieved at low to moderate concentrations.

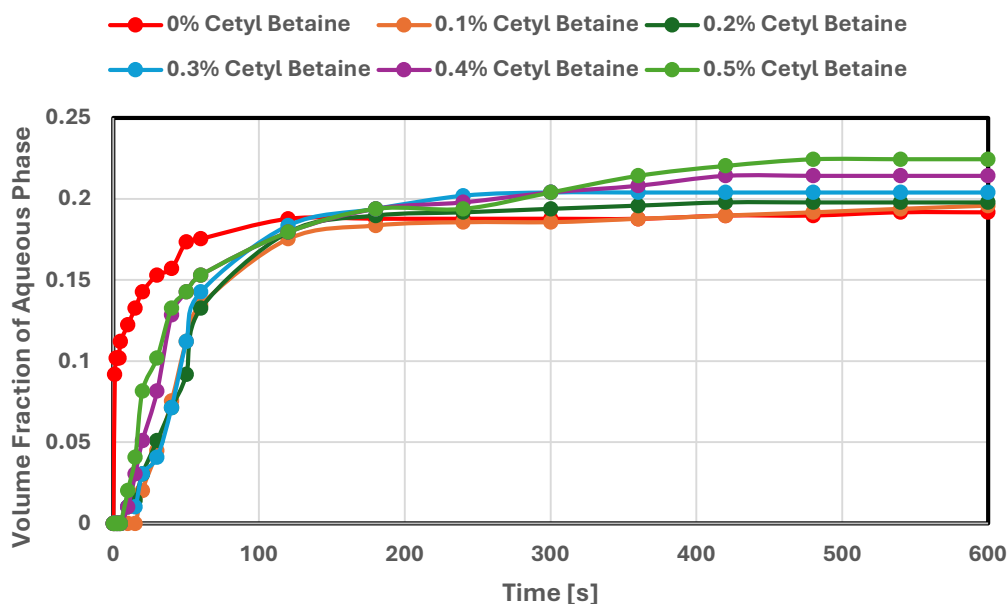


Figure 32. Coalescence stability of W/O emulsion of oil and cetyl betaine solution with varying cetyl betaine concentration.

4.4.6. Comparative Analysis of Coalescence Stability

NCC-stabilized emulsions exhibited markedly improved coalescence resistance compared with the control, showing strong stability even at low concentrations. The results indicate that coalescence was delayed across all NCC loadings, confirming effective stabilization at relatively small additions. Beyond approximately 0.2 wt%, the extent of coalescence remained nearly constant, suggesting that the interface had reached saturation and that further increases in NCC concentration provided little additional benefit.

Surfactant-stabilized emulsions displayed distinct concentration-dependent behaviors depending on surfactant type. The most diverse profile was observed for SDS, where the lowest concentration (0.05 wt%) produced the highest coalescence stability, followed by an increase in coalescence rate up to 0.3 wt%, and then a slight reduction at higher concentrations, reflecting complex interfacial dynamics. Nonetheless, all SDS formulations showed improved stability relative to the control. OTAC also enhanced coalescence stability, with similar improvements observed up to 0.3 wt%; above this concentration, greater emulsification resulted in an increased apparent aqueous phase volume due to the formation of a thick, cream-like layer at the phase boundary.

The alcohol ethoxylate system (Alfonic® C12–14 ethoxylate) exhibited gradual improvement in coalescence resistance with increasing concentration, with higher loadings contributing to slower separation rates and more persistent emulsions. In contrast, the cetyl betaine (Amphosol®) emulsions showed the opposite trend, where higher concentrations led to faster coalescence rates. Despite these differences, all surfactant systems demonstrated greater coalescence stability than the control, confirming that each provided measurable stabilization across the tested concentration ranges.

Chapter 5. Discussion

5.1. Mechanistic Insights

The results presented in Chapter 4 collectively highlight the contrasting stabilization mechanisms governing emulsions stabilized by NCC and those stabilized by molecular surfactants. While both systems achieved measurable improvements in emulsion stability relative to the control, their underlying mechanisms differ fundamentally. The differences observed in interfacial tension, phase inversion behavior, and coalescence stability are consistent with two distinct modes of interfacial stabilization: dynamic surfactant-driven adsorption and static particle-driven jamming. These mechanisms not only govern the kinetics of droplet formation and coalescence but also determine how each system responds to increasing internal phase fraction and prolonged storage.

5.1.1. Surfactant-Driven versus Particle-Driven Stabilization

Surfactant-stabilized emulsions rely primarily on the rapid adsorption and desorption of amphiphilic molecules at the oil–water interface. The reduction in interfacial tension achieved by SDS, OTAC, alcohol ethoxylate, and cetyl betaine facilitated droplet breakup during emulsification and temporarily stabilized the dispersed phase. As seen in Section 4.3, the equilibrium interfacial tension decreased sharply at low concentrations for all surfactants and reached a plateau once the interface became saturated. This dynamic adsorption allows surfactants to continuously rearrange in response to deformation or coalescence, lending them adaptability but also limiting their long-term stability. Once surfactant molecules desorb or redistribute under shear or diffusion, the interface becomes vulnerable to film rupture and droplet coalescence.

In contrast, NCC-stabilized emulsions exhibited behavior characteristic of Pickering-type stabilization. The particulate nature of NCC results in irreversible adsorption at the oil–water interface, where the particles anchor through partial wetting and assemble into rigid, jammed layers. These layers act as mechanical barriers that suppress film drainage and droplet deformation. The minimal change in surface tension across NCC concentrations (Section 4.3) confirms that stabilization is not driven by interfacial energy reduction but by steric and mechanical reinforcement. Once sufficient particles accumulate at the interface, a percolating network forms that physically arrests coalescence, leading to the high long-term stability observed in the

coalescence tests. The concentration-independent behavior above approximately 0.2 wt% further supports the conclusion that NCC stabilization depends on structural coverage rather than dynamic equilibration.

Overall, the surfactant-driven systems are governed by kinetic adsorption–desorption equilibria, whereas NCC-driven systems are governed by interfacial crowding and rigidity. Surfactant stabilization is dynamic and reversible; particle stabilization is static and irreversible. This fundamental difference explains the delayed catastrophic inversion and superior coalescence resistance observed for NCC compared with molecular surfactants.

5.1.2. Role of Interfacial Jamming, Surface Tension Reduction, and Steric Hindrance

The distinct physical origins of stabilization are further clarified by considering three interfacial mechanisms: interfacial jamming, surface tension reduction, and steric hindrance, all of which manifest differently in particle- versus surfactant-stabilized systems.

In the case of surfactants, the dominant effect is surface tension reduction, which lowers the energy barrier to droplet formation and temporarily stabilizes the interface. The surface and interfacial tension results (Section 4.3) showed steep declines with increasing surfactant concentration, particularly for alcohol ethoxylate and cetyl betaine systems, confirming efficient molecular packing at the interface. However, once saturation was achieved, excess surfactant accumulated in the bulk phase [15]. This redistribution promotes transient stabilization rather than structural reinforcement, explaining why coalescence resistance decreased or plateaued at higher concentrations.

For NCC, interfacial jamming and steric hindrance dominate. The rod-like geometry and amphiphilic surface chemistry of NCC allow for irreversible adsorption, followed by lateral packing that generates a mechanically stiff interfacial layer[76,154,160]. As the surface becomes crowded, further particle movement is restricted, and the interface transitions into a jammed state. This process produces long lasting emulsions with minimal coalescence and no significant change in interfacial tension beyond 0.2 wt%. The resulting stability arises from a balance between excluded volume effects and interparticle interactions rather than from energy minimization. Steric

hindrance introduced by protruding cellulose rods further prevents close approach of droplets, effectively halting film drainage and suppressing coalescence even under mild agitation or extended storage.

5.2. Theoretical Frameworks in Light of Findings

The findings of this study can be further understood through established theoretical frameworks that describe emulsion type, curvature, and inversion phenomena. In particular, Bancroft's rule, the hydrophilic–lipophilic deviation (HLD) theory, and catastrophic inversion models provide complementary perspectives for interpreting the observed differences between surfactant- and particle-stabilized emulsions. Although these frameworks were originally developed for molecular surfactants, their comparison with NCC behavior helps reveal how particle-stabilized systems diverge from traditional emulsion theory.

5.2.1. Bancroft's Rule and Curvature Preference

According to Bancroft's rule, the continuous phase of an emulsion depends on the solubility of the emulsifier, with hydrophilic surfactants favoring oil-in-water (O/W) systems and lipophilic ones stabilizing water-in-oil (W/O) emulsions. In this study, all four surfactants—SDS, OTAC, alcohol ethoxylate, and cetyl betaine—are hydrophilic in nature and would therefore be expected to produce O/W emulsions. However, this prediction did not hold at low concentrations, where all systems initially stabilized as W/O emulsions. Bancroft's rule became evident only after the surfactant concentration increased beyond a certain threshold, at which point the emulsions progressively inverted toward O/W structures. This concentration-dependent transition indicates that at low levels, the limited surfactant coverage allows greater solubility and partitioning into the oil phase, favoring W/O stabilization. As concentration increases, the enhanced adsorption of surfactant molecules at the interface promotes curvature toward the aqueous phase, leading to inversion behavior that aligns with Bancroft's rule. The strong dependence of inversion point on surfactant concentration reflects the surfactant's ability to redistribute between oil and water phases in response to curvature changes, consistent with classical emulsification behavior.

However, the NCC-stabilized emulsions deviate markedly from this rule. Despite the hydrophilic surface chemistry of NCC, these systems successfully stabilized W/O emulsions across a broad range of internal phase fractions without inversion. This behavior cannot be explained solely by solubility or curvature preference. Instead, NCC acts as a solid amphiphile where the adsorption is determined by particle wettability and aspect ratio rather than molecular solubility. Once adsorbed, NCC forms rigid interfacial layers that lock in the emulsion structure, effectively

overriding the curvature-driven tendencies predicted by Bancroft's rule. This demonstrates that while Bancroft's rule remains valid for molecular surfactants, it is not directly applicable to Pickering systems where the emulsifier becomes an immobile structural component of the interface.

5.2.2. HLD and HLD–NAC Frameworks in Context

Although the hydrophilic–lipophilic deviation (HLD) and hydrophilic–lipophilic deviation–net average curvature (HLD–NAC) frameworks are traditionally applied to surfactant systems where salinity, oil type, and temperature are tunable, their conceptual principles remain relevant for interpreting the curvature and stability trends observed in this study. In the absence of explicit salinity or compositional variation, the role of surfactant concentration itself can be viewed as an internal variable that alters interfacial curvature and effective HLD.

In the classical HLD formulation, a positive value indicates a tendency toward water-in-oil (W/O) emulsions, while a negative value favors oil-in-water (O/W) structures, with $HLD = 0$ representing a balanced or bicontinuous state. Within this conceptual framework, the behavior of the surfactant systems observed here aligns with expected curvature tendencies. At low concentrations, all surfactants exhibited reduced interfacial tension and delayed inversion, corresponding to an approach toward the balanced ($HLD \approx 0$) condition where interfacial curvature is most stable. As concentration increased, the interfaces became saturated and micellization in the aqueous phase likely shifted the effective curvature toward the O/W direction ($HLD < 0$), consistent with earlier inversion points and faster coalescence. The distinct responses among surfactants i.e. sharper curvature shifts for SDS, OTAC, and cetyl betaine and more stable behavior for alcohol ethoxylate reflect differences in headgroup charge and chain packing, which conceptually parallel changes in HLD parameters such as characteristic length and hydrophilic strength.

The HLD–NAC framework extends these ideas by incorporating geometric factors such as interfacial elasticity and curvature flexibility. Although explicit NAC parameters were not calculated, the experimental results qualitatively support its central premise: interfacial stability depends on how readily surfactant layers accommodate curvature as the internal phase fraction increases. Surfactants with flexible interfacial films, such as alcohol ethoxylate, maintained stability over a wider concentration range, whereas those with higher charge density or dipolar

headgroups, such as SDS and cetyl betaine, exhibited sharper transitions once the interface could no longer adjust.

However, the behavior of NCC falls largely outside the scope of the HLD and HLD–NAC frameworks. Unlike molecular surfactants, NCC does not dynamically tune interfacial curvature through adsorption–desorption equilibria.

In this light, the HLD and HLD–NAC models remain valuable as conceptual tools for interpreting the surfactant systems studied here, providing insight into how concentration-dependent curvature and interfacial flexibility influence stability. Yet, their limitations also highlight the need for expanded theoretical frameworks that can capture particle-induced rigidity and the mechanically dominated stabilization observed in Pickering emulsions.

5.2.3. Catastrophic Phase Inversion Models

Catastrophic phase inversion models describe the abrupt transition from W/O to O/W emulsions as the dispersed phase volume fraction increases and the emulsion crosses the close-packing limit. For surfactant-stabilized systems, this transition is strongly influenced by interfacial mobility and coalescence dynamics. The conductivity data presented in Section 4.1 showed that the inversion point decreased with increasing surfactant concentration once the interface became saturated. This behavior aligns with kinetic models proposed by Salager and Forgiarini [130], in which inversion is triggered when droplet crowding, and surfactant redistribution can no longer sustain continuous-phase connectivity.

5.3. Implications for Industrial Applications

The contrasting behaviors of surfactant and NCC stabilized emulsions have important implications for how emulsions are designed and applied in industrial processes. The delayed catastrophic inversion, shown by the increase in the critical aqueous phase volume fraction from 0.25 in the control to 0.55 at 1.5 wt% NCC, reflects the enhanced stability of NCC systems and their shift from dynamic, surfactant-controlled interfaces to mechanically reinforced particle-stabilized structures. This shift fundamentally alters how emulsions respond to high stress, variable salinity, and extended storage, which are common challenges in industrial operations.

In high internal phase emulsions (HIPEs), where the dispersed phase volume fraction typically exceeds 0.74, the random close packing limit of spheres, droplets deform and jam into a polyhedral network that gives the system a solid-like character. The delayed inversion observed with NCC suggests that particle-stabilized emulsions could sustain very high internal phase ratios without coalescence or collapse of the continuous phase. This ability to maintain structure at high internal phase fractions makes NCC-stabilized systems attractive as templates for porous polymers, structural foams, and coatings where control of microstructure is essential. The nonlinear increase in the critical inversion threshold, which rises rapidly below 0.5 wt% NCC and then gradually levels off, points to a transition from partial coverage to interfacial saturation. Beyond this point, the particle layer reaches its maximum interfacial packing, suggesting that varying NCC concentration can be used to control the rigidity and structural stability of HIPE systems.

In enhanced oil recovery (EOR), the differences in inversion behavior and interfacial tension reduction translate directly to process performance. Surfactant stabilized emulsions invert at relatively low aqueous fractions as interfacial tension falls below about 5 mN/m, reflecting highly mobile oil–water interfaces that promote phase inversion. Their stability is also strongly influenced by salinity. As the ionic strength of the aqueous phase increases, ions compress the electrical double layer surrounding droplets, reducing the Debye length, and weakening electrostatic repulsion. According to DLVO theory, this reduction in the repulsive barrier allows van der Waals attraction to dominate, promoting droplet coalescence and destabilization [1,82]. Consequently, surfactant stabilized emulsions often lose stability or invert prematurely under high salinity or when process water is used instead of purified water.

NCC, on the other hand, maintained a nearly constant interfacial tension near 38 mN/m across all concentrations, yet the emulsions remained stable and resisted inversion even at high water loadings. This independence from interfacial tension reduction and ionic screening indicates that NCC stabilization is governed by mechanical and steric effects rather than by molecular adsorption dynamics. Because the interfacial layer is formed through irreversible particle attachment and physical jamming, NCC-stabilized emulsions are far less sensitive to changes in ionic strength or electrolyte composition. Such behavior suggests that NCC-stabilized emulsions would perform reliably under variable salinity and shear conditions, where surfactant systems typically fail. These characteristics can lower chemical cost and environmental burden while improving operational reliability in demanding field environments.

The same stabilization advantages are relevant for food, cosmetics, and pharmaceutical formulations where long-term stability and biocompatibility are crucial. The coalescence results showed that even low NCC loadings of 0.2 wt% significantly reduced phase separation over time, achieving steady stability comparable to that of optimized surfactant systems. Unlike ionic surfactants, which exhibited an optimal range followed by reduced performance at higher concentrations, NCC maintained consistent and durable stabilization across all concentrations. Because NCC stabilization does not depend on pH or ionic strength, it can operate effectively in a wide range of formulations including those with electrolytes or low water activity, such as creams, ointments, and encapsulation systems.

Overall, these results show that NCC-stabilized Pickering emulsions offer both functional and environmental advantages over conventional surfactant systems. The shift from surfactant to particle-based stabilization provides a sustainable and reliable approach for achieving stable performance across diverse industrial conditions, from polymer templating and energy recovery to consumer and pharmaceutical applications.

5.4. Sustainability Considerations

The use of NCC as an emulsion stabilizer offers clear sustainability benefits that extend beyond formulation performance. Derived from renewable cellulose feedstocks, NCC is biodegradable, non-toxic, and produced through processes with significantly lower carbon intensity than the synthesis of petroleum-based surfactants. Its sourcing from forestry residues and other biomass streams supports circular economy principles by transforming low-value by-products into high-function materials. From an industrial perspective, the replacement of synthetic surfactants with NCC can directly contribute to carbon reduction targets and corporate sustainability metrics tied to greenhouse gas emissions and resource efficiency.

Reducing dependence on synthetic surfactants also aligns with increasingly strict environmental regulations governing effluent quality, aquatic toxicity, and surfactant biodegradability. Many traditional surfactants persist in the environment, contribute to chemical oxygen demand, and require energy-intensive wastewater treatment. NCC, in contrast, degrades naturally and introduces no secondary pollutants, allowing for easier compliance with regulatory frameworks such as REACH and OSPAR that promote the use of inherently biodegradable substances. Its stability across broad pH and salinity ranges further decreases the need for multiple formulation additives, reducing material complexity and life-cycle impact.

In this context, the transition from surfactant to particle-based stabilization represents more than a change in formulation strategy; it is a shift toward sustainable process design. By integrating NCC as a renewable and low-impact stabilizer, industries can reduce chemical load, extend product lifetime, and improve environmental performance while meeting both regulatory and operational objectives.

5.5. Limitations of this Study

While this work provides new insight into the contrasting phase inversion behaviors of surfactants and NCC-stabilized emulsions, several limitations should be acknowledged when interpreting the findings. First, droplet size distribution was not directly measured, and all comparisons were made under the assumption of similar shear and mixing conditions. Although care was taken to maintain consistent homogenization parameters, variations in droplet size or polydispersity could influence the onset of catastrophic inversion and stability trends. Future work should therefore incorporate droplet size analysis to quantitatively link interfacial phenomena with emulsion microstructure.

Second, interfacial rheology was not evaluated, which limits the mechanistic interpretation of how interfacial viscoelasticity contributes to the observed differences in inversion and coalescence stability. Measurements of interfacial storage and loss moduli would help clarify the extent to which mechanical rigidity, jamming, or relaxation processes govern the transition from dynamic to arrested interfacial behavior in NCC stabilized systems.

Finally, the experiments were conducted under controlled laboratory conditions using a well-defined model oil–water system. While this approach isolates fundamental mechanisms, it does not fully capture the chemical and mechanical complexity of industrial emulsions, which often contain co-solvents, surfactant mixtures, or particulate impurities. Extending this work to more representative systems and flow conditions will be necessary to validate the practical applicability of NCC as a stabilizer under realistic processing and field environments.

5.6. Future Work

Building on the limitations identified in this study, several directions can be pursued to advance the understanding of catastrophic phase inversion in NCC stabilized emulsions. Future work should incorporate interfacial rheology measurements to quantify the viscoelastic properties of the NCC layer and clarify how interfacial elasticity, storage modulus, and relaxation behavior influence the resistance to inversion. These measurements would also allow direct comparison with surfactant-stabilized systems, where interfacial mobility and dynamic adsorption are dominant.

Investigating the role of droplet polydispersity and size evolution under controlled shear would further strengthen the mechanistic framework developed here. Quantitative droplet size data would enable correlation of interfacial properties with inversion kinetics and coalescence behavior, particularly under variable shear or temperature conditions.

Another promising direction involves hybrid NCC–surfactant systems, where controlled co-adsorption could yield synergistic stabilization effects. Such combinations may enhance interfacial packing density, modify curvature, or produce tunable inversion thresholds, bridging the gap between classical surfactant control and particle induced rigidity. Exploring these hybrid interfaces under industrially relevant formulations and salinity conditions will be key to expanding the practical applicability of NCC in complex multiphase systems.

Chapter 6. Conclusions

This work examined the catastrophic phase inversion behavior of emulsions stabilized by nanocrystalline cellulose (NCC) and compared it with emulsions stabilized by conventional molecular surfactants of differing headgroup charge types. By integrating conductivity measurements, interfacial tension data, and coalescence stability analysis, the study established a quantitative and mechanistic understanding of how NCC and surfactants govern phase inversion through fundamentally distinct pathways. The findings represent one of the first systematic investigations of water-in-oil to oil-in-water inversion using NCC under controlled conditions and provide new insight into how particle-driven stabilization reshapes the inversion landscape traditionally described by surfactant-based frameworks.

6.1. Summary of Findings

The results demonstrated that NCC markedly delays catastrophic phase inversion compared with surfactant-stabilized systems. The critical aqueous phase volume fraction required for inversion increased from 0.25 in the surfactant-free control to 0.55 at 1.5 wt% NCC, indicating a nonlinear relationship between NCC concentration and inversion threshold. This trend reflects a transition from limited particle coverage at low concentrations to interfacial saturation and jamming at higher loadings, where the droplet interfaces become mechanically constrained and resist structural rearrangement.

In contrast, surfactant-stabilized systems exhibited non-monotonic inversion behavior. At low surfactant concentrations, inversion was delayed due to limited interfacial coverage and kinetic stabilization of the dispersed phase. At higher concentrations, however, inversion occurred at lower aqueous phase fractions as the surfactant rapidly redistributed and saturated the interface, lowering interfacial tension to values below approximately 5 mN/m. This transition from delayed to premature inversion highlights the dynamic and reversible nature of surfactant adsorption and its sensitivity to concentration and interfacial curvature.

Interfacial and surface tension measurements reinforced these mechanistic distinctions. NCC exhibited minimal surface activity, maintaining an interfacial tension near 38 mN/m across all concentrations tested, whereas all surfactants produced steep reductions in both surface and interfacial tension, confirming their molecular mobility and dynamic interfacial exchange. Despite its weak surface activity, NCC imparted superior coalescence resistance above 0.2 wt%, where irreversible particle adsorption and steric hindrance formed mechanically rigid interfaces. Collectively, these results support the view that NCC stabilization is governed by mechanical and structural effects rather than by thermodynamic minimization of interfacial energy.

Coalescence stability measurements further demonstrated that NCC produced durable emulsions with minimal phase separation over time, while surfactant-stabilized emulsions exhibited variable stability depending on concentration and surfactant class. These findings confirm that catastrophic inversion and coalescence stability are not independent but interconnected through interfacial mechanics. The delayed inversion and reduced coalescence observed for NCC can both be attributed to interfacial reinforcement through particle jamming, which maintains droplet integrity even as dispersed-phase volume increases.

6.2. Advances in Knowledge

This study contributes several new advances to the understanding of catastrophic phase inversion and emulsion stabilization.

First, it provides the first quantitative evidence that NCC can systematically delay inversion through a concentration-dependent mechanism linked to interfacial packing density. While previous studies had established NCC's ability to stabilize emulsions, its specific influence on catastrophic inversion had not been measured under controlled, comparable conditions. The progressive shift in inversion point observed here establishes a direct experimental relationship between particle concentration, interfacial coverage, and inversion resistance.

Second, the results clarify the mechanistic divergence between particle- and surfactant-stabilized systems. In surfactant emulsions, inversion is a kinetically driven process where curvature adaptation, micellar dynamics, and interfacial saturation dictate the transition point. In NCC emulsions, inversion is constrained by irreversible adsorption and mechanical rigidity, resulting in a fundamentally different inversion pathway that proceeds only when geometric packing limits are reached. This finding bridges interfacial phenomena with catastrophic inversion theory, extending existing models such as Bancroft's rule and hydrophilic-lipophilic deviation (HLD) theory beyond molecular surfactants to include particulate stabilizers (or capture the deviation away from these models).

Third, the study reveals the limitations of interfacial tension as a predictive parameter for inversion in particle-stabilized systems. While surfactant inversion correlates with minima in interfacial tension, NCC stabilization proceeds independently of interfacial energy reduction. This decoupling highlights the need for new theoretical frameworks that incorporate interfacial elasticity and steric constraints alongside classical curvature-based models. The results therefore, advance both conceptual and practical understanding of how interfacial mechanics influence emulsion inversion.

Finally, the comparison across surfactants with different charge types—anionic (SDS), cationic (OTAC), nonionic (Alfonic®, C12-C14 alcohol ethoxylate), and zwitterionic (Amphosol®, cetyl betaine)—demonstrates that the inversion response is not only specific to charge class but instead governed by interfacial coverage and dynamic redistribution. This observation supports a unifying

kinetic view of surfactant-driven inversion while highlighting the uniqueness of NCC's mechanical stabilization.

6.3. Practical and Industrial Relevance

Beyond scientific insight, the findings carry broad implications for emulsion design across industrial sectors. NCC's ability to stabilize emulsions through physical jamming rather than electrostatic or molecular adsorption offers practical advantages in processes where ionic composition, temperature, or shear conditions fluctuate.

In high internal phase emulsions (HIPEs), NCC can maintain structural integrity at internal phase fractions that would otherwise induce inversion or collapse in surfactant systems. This property is valuable for producing porous polymeric materials, foams, and coatings that rely on stable, high-volume emulsions as templates for microstructure formation.

In enhanced oil recovery (EOR) and other subsurface processes, NCC's insensitivity to salinity overcomes a major limitation of ionic surfactants. According to DLVO theory, increased salinity compresses the electrical double layer and reduces the Debye length, weakening repulsive forces that stabilize surfactant-stabilized droplets. NCC, however, maintains stability through steric and mechanical means, making it a promising alternative in reservoirs with variable salinity and temperature. These characteristics not only improve process reliability but can also reduce the chemical cost and environmental burden associated with surfactant loss and brine treatment.

In food, cosmetic, and pharmaceutical formulations, NCC provides biocompatible and non-toxic stabilization, extending product shelf life without the need for synthetic surfactants. Its renewable and biodegradable nature aligns with increasing regulatory pressure to minimize persistent organic ingredients in consumer products. Furthermore, its ability to maintain stable emulsions across wide pH and ionic conditions simplifies formulation development and broadens applicability across multiple product types.

At a broader level, replacing synthetic surfactants with NCC contributes to corporate sustainability and carbon reduction goals by reducing dependency on petrochemical inputs and improving life-cycle performance. NCC's derivation from renewable biomass and compatibility with existing processing technologies make it a practical and scalable option for industries seeking both technical and environmental performance improvements.

6.4. Closing Remarks

Overall, this study establishes that NCC is not merely a renewable substitute for molecular surfactants but a fundamentally different class of stabilizer that governs catastrophic phase inversion through mechanical constraint rather than interfacial thermodynamics. By quantifying its stabilizing influence and identifying the conditions under which it delays inversion, this work bridges the gap between classical surfactant science and emerging particle-based emulsion stabilization. The mechanistic and practical insights gained here provide a foundation for designing stable, sustainable emulsions that perform reliably across diverse industrial environments.

Looking ahead, the insights gained from this work can inform both theoretical modeling and applied formulation strategies that extend beyond laboratory-scale systems. By coupling future interfacial rheology and droplet-scale analyses with process-level validation, the principles established here can guide the development of next-generation emulsions that unite performance, stability, and sustainability across industrial, environmental, and biomedical applications.

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Appendix – Raw Data

i. Phase Inversion

NCC – Phase Inversion

0% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	0
800	0.049	40.8	0.04	N	40
800	0.072	61.8	0.07	N	45
800	0.082	71.1	0.07	N	50
800	0.112	100.8	0.31	N	50
800	0.136	126.2	0.99	N	50
800	0.162	154.6	17.05	N	55
800	0.192	190	22.5	N	60
800	0.210	213.2	32.5	N	55
800	0.234	244.3	44.2	N	55
800	0.252	269.4	240.1	Y	60
800	0.267	291.4	260.5	Y	55
800	0.290	327	290.6	Y	55
800	0.316	369.6	323	Y	50
800	0.334	401.2	346	Y	50
800	0.357	444	375	Y	50
800	0.383	496.3	410	Y	50
800	0.396	525.3	427	Y	50
800	0.415	568.4	453	Y	50
800	0.439	626.9	478	Y	50
800	0.457	674.6	498	Y	50
800	0.477	729	523	Y	50
800	0.490	767.6	540	Y	50
800	0.502	805.1	556	Y	50
800	0.512	840.3	570	Y	50
800	0.524	882.3	587	Y	50
800	0.540	940	602	Y	50
800	0.550	977.3	620	Y	50
800	0.562	1025.7	633	Y	50
800	0.572	1068.9	643	Y	50
800	0.583	1118.2	658	Y	50
800	0.602	1207.6	686	Y	50

0.10% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.038	31.9	0	N	35
800	0.064	55.1	0.43	N	48
800	0.093	81.7	0.3	N	48
800	0.110	98.8	0.38	N	50
800	0.116	105.3	0.39	N	50
800	0.143	133.7	0.52	N	50
800	0.153	144.4	0.79	N	50
800	0.166	159.3	1.23	N	50
800	0.178	173.6	2.03	N	50
800	0.190	187.1	4.11	N	50
800	0.205	206	5.56	N	52
800	0.213	215.9	8.83	N	52
800	0.220	225.4	9.22	N	52
800	0.230	238.8	8.26	N	50
800	0.242	255.1	9.11	N	50
800	0.250	266.8	18.68	N	50
800	0.259	279.3	18.08	N	50
800	0.266	289.8	21.61	N	50
800	0.277	306.9	21.9	N	50
800	0.283	315.9	20.47	N	50
800	0.289	324.7	23.88	N	50
800	0.297	338.3	27.32	N	50
800	0.311	360.9	31	N	50
800	0.315	368.5	42	N	50
800	0.323	381.4	340	Y	50
800	0.332	398	360	Y	40
800	0.346	423.4	380	Y	40
800	0.365	459.1	405	Y	40
800	0.377	484.3	421	Y	40
800	0.387	506	435	Y	40
800	0.399	531.1	450	Y	40
800	0.413	563.8	470	Y	40
800	0.433	609.8	494	Y	40
800	0.443	635	506	Y	40
800	0.454	665.6	519	Y	40
800	0.465	694.7	532	Y	40
800	0.478	731.5	547	Y	40
800	0.485	754.7	557	Y	40
800	0.500	801.2	575	Y	40

800	0.516	853.7	595	Y	40
800	0.535	919.8	617	Y	40
800	0.543	951.7	627	Y	40
800	0.564	1033.8	654	Y	40
800	0.581	1110.1	676	Y	40

0.20% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.052	43.9	0.03	N	40
800	0.096	85	0.06	N	50
800	0.123	112.1	1.81	N	50
800	0.147	137.5	1.29	N	55
800	0.159	150.7	1.06	N	52
800	0.178	173	1.27	N	52
800	0.192	189.6	1.38	N	52
800	0.207	208.6	2.14	N	52
800	0.225	232	2.65	N	52
800	0.242	254.9	3.07	N	52
800	0.252	269.9	3.63	N	52
800	0.262	284.3	4.53	N	52
800	0.273	299.9	4.86	N	52
800	0.284	318	6.24	N	52
800	0.290	327	7.69	N	52
800	0.298	339	8.3	N	52
800	0.306	352	10.45	N	52
800	0.312	362.4	11.9	N	52
800	0.319	374.6	13.55	N	52
800	0.329	391.7	18.79	N	52
800	0.334	401.8	22.25	N	52
800	0.341	414.7	24.9	N	52
800	0.350	430.4	24.15	N	52
800	0.357	444.7	20.77	N	49
800	0.368	465.3	22.92	N	49
800	0.376	481.6	19.8	N	49
800	0.386	502.3	21.15	N	49
800	0.393	517.3	22.91	N	49
800	0.403	540.2	512	N	49
800	0.414	565.9	546	Y	49
800	0.423	586.9	565	Y	49
800	0.436	618.8	582	Y	49

800	0.447	646	597	Y	49
800	0.464	692.7	620	Y	49
800	0.477	730	637	Y	49
800	0.500	799.4	667	Y	49
800	0.521	871.1	696	Y	49
800	0.541	944.7	721	Y	49
800	0.560	1018	744	Y	49
800	0.574	1077.6	762	Y	49
800	0.591	1155.4	782	Y	49

0.30% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.073	63.2	0.58	N	40
800	0.104	92.6	0.98	N	40
800	0.128	117.9	0.7	N	45
800	0.157	149.3	0.81	N	45
800	0.178	173.4	1.63	N	50
800	0.195	193.6	1.48	N	50
800	0.225	231.9	1.99	N	50
800	0.235	246.2	1.83	N	50
800	0.246	261.2	2.11	N	50
800	0.254	271.8	2.13	N	50
800	0.265	287.8	2.5	N	50
800	0.272	298.4	2.78	N	50
800	0.280	311.6	2.22	N	50
800	0.292	329.2	2.34	N	50
800	0.302	345.4	2.62	N	50
800	0.313	364	2.64	N	50
800	0.322	380.8	2.77	N	50
800	0.330	394.3	2.83	N	50
800	0.341	414.5	3.03	N	50
800	0.352	435.4	3.71	N	50
800	0.367	463.8	3.79	N	50
800	0.374	477	4.06	N	50
800	0.381	492.6	5.18	N	50
800	0.388	506.6	5.52	N	50
800	0.395	521.8	6.1	N	52
800	0.400	534	7.36	N	52
800	0.408	550.4	8.6	N	52
800	0.414	565.9	9.7	N	52

800	0.421	581.3	10.68	N	52
800	0.427	597.2	12.23	N	52
800	0.436	618.2	576	Y	52
800	0.451	656	603	Y	52
800	0.468	702.4	633	Y	49
800	0.485	753.8	655	Y	49
800	0.503	809.2	678	Y	49
800	0.516	853.7	696	Y	49
800	0.539	933.8	722	Y	49
800	0.563	1029	750	Y	49
800	0.582	1115.7	772	Y	49
800	0.599	1195.9	790	Y	49

0.50% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.108	97.1	0	N	40
800	0.120	109.5	0.59	N	45
800	0.135	125.3	0.5	N	45
800	0.154	145.3	0.66	N	50
800	0.174	168.9	1.03	N	50
800	0.187	184	0.99	N	50
800	0.204	204.6	0.9	N	50
800	0.226	233.3	1.07	N	50
800	0.238	249.8	1.23	N	50
800	0.259	278.9	1.55	N	50
800	0.270	296	1.52	N	50
800	0.292	329.6	1.64	N	50
800	0.302	346.4	1.91	N	50
800	0.316	369	2.07	N	50
800	0.324	384.1	2.22	N	50
800	0.334	400.7	2.38	N	50
800	0.342	415	2.64	N	50
800	0.350	430	2.04	N	50
800	0.360	450.1	2.06	N	50
800	0.368	465.6	2.09	N	50
800	0.380	490.9	2.29	N	50
800	0.387	504.7	2.54	N	50
800	0.393	518.5	2.71	N	50
800	0.402	538.5	2.87	N	50
800	0.408	551.9	3.12	N	50

800	0.414	566	3.25	N	50
800	0.419	577.9	3.73	N	50
800	0.426	592.9	4.03	N	50
800	0.435	616.5	4.21	N	50
800	0.442	633.7	4.48	N	52
800	0.448	648.5	5.06	N	55
800	0.458	675.5	6	N	55
800	0.464	691.2	6.96	N	55
800	0.468	703.8	677	Y	55
800	0.477	730.8	694	Y	55
800	0.493	777.5	717	Y	50
800	0.503	808.8	733	Y	45
800	0.517	857.4	752	Y	45
800	0.537	929.5	774	Y	45
800	0.560	1019.6	801	Y	45
800	0.587	1136.1	829	Y	45
800	0.600	1199.8	844	Y	45

0.70% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	52.5	0.09	N	30
800	0.094	83.2	0.11	N	40
800	0.143	133.4	0.24	N	45
800	0.159	151.6	0.34	N	45
800	0.177	171.6	0.41	N	50
800	0.195	194	0.57	N	50
800	0.214	217.8	0.6	N	50
800	0.233	243.3	0.75	N	50
800	0.253	270.3	0.82	N	50
800	0.267	290.8	0.88	N	50
800	0.283	315.6	1.02	N	50
800	0.297	338.2	1.23	N	50
800	0.310	360	1.34	N	50
800	0.329	393	1.54	N	50
800	0.342	416.1	1.84	N	50
800	0.351	432.3	2.04	N	50
800	0.364	458.5	2.23	N	50
800	0.377	485	2.34	N	50
800	0.392	515.5	2.47	N	50
800	0.399	530.1	2.7	N	50
800	0.405	544.3	2.86	N	50

800	0.412	560.6	3.13	N	50
800	0.419	577.8	3.35	N	50
800	0.423	586.6	3.45	N	50
800	0.427	595.3	3.62	N	50
800	0.432	608.7	3.87	N	50
800	0.436	617.5	4.13	N	50
800	0.440	629.1	4.58	N	50
800	0.446	643.9	5.12	N	52
800	0.451	656.3	6.17	N	53
800	0.454	666.4	6.85	N	53
800	0.459	678.9	7.37	N	53
800	0.463	689.1	8.15	N	53
800	0.466	697.5	9.68	N	53
800	0.469	705.2	11.67	N	53
800	0.471	712.8	708	Y	53
800	0.480	737.9	724	Y	45
800	0.491	771.7	738	Y	45
800	0.508	825.8	757	Y	45
800	0.521	869.8	772	Y	45
800	0.534	916.3	787	Y	45
800	0.548	971.7	803	Y	45
800	0.564	1035.9	822	Y	45
800	0.579	1100.6	838	Y	45
800	0.599	1195.5	860	Y	45
800	0.601	1205.7	863	Y	45

0.90% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.061	52.1	0	N	40
800	0.083	72.3	0.28	N	45
800	0.112	100.8	0.39	N	50
800	0.149	139.6	0.88	N	50
800	0.173	167.6	0.61	N	50
800	0.199	199.3	0.78	N	55
800	0.219	224.7	0.81	N	55
800	0.252	269.6	1.12	N	55
800	0.268	292.2	1.25	N	55
800	0.286	320.8	1.71	N	55
800	0.301	344.5	3.46	N	60
800	0.319	374	5.5	N	60

800	0.336	405	6.11	N	60
800	0.353	436.3	29.8	N	60
800	0.366	461.1	32.8	N	60
800	0.378	486.4	34.5	N	60
800	0.402	537	35.8	N	60
800	0.409	554.7	32.7	N	60
800	0.421	581.1	12.9	N	60
800	0.431	605.6	8.54	N	60
800	0.439	626.8	9.52	N	60
800	0.447	647.2	12.15	N	60
800	0.454	664.4	15.14	N	60
800	0.463	689.7	25.86	N	60
800	0.470	710.1	28.2	N	60
800	0.476	728.1	38.6	N	60
800	0.483	746.4	797	Y	40
800	0.504	814.5	825	Y	40
800	0.521	870.5	843	Y	40
800	0.539	937.1	862	Y	40
800	0.546	960.6	867	Y	40
800	0.549	975.6	869	Y	40
800	0.555	995.9	874	Y	40
800	0.560	1017.4	876	Y	40
800	0.569	1057.8	886	Y	40
800	0.575	1082.9	889	Y	40
800	0.581	1111.2	895	Y	40
800	0.590	1152	906	Y	40
800	0.599	1193.4	914	Y	40
800	0.604	1221.9	918	Y	40

1.10% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.072	62.2	0.11	N	30
800	0.110	98.5	0.13	N	40
800	0.139	129	0.25	N	45
800	0.160	152.7	0.36	N	50
800	0.187	184.1	0.54	N	50
800	0.208	210.4	0.78	N	50
800	0.228	236.9	0.97	N	55
800	0.245	259.7	1.26	N	55
800	0.271	297.3	1.16	N	55

800	0.286	320.1	1.6	N	55
800	0.299	341.3	1.77	N	55
800	0.313	364.8	1.86	N	55
800	0.330	393.6	1.96	N	55
800	0.345	421.8	2.23	N	55
800	0.361	452	2.31	N	55
800	0.374	478.4	2.47	N	55
800	0.386	502.9	2.51	N	55
800	0.395	522	2.89	N	55
800	0.406	546.9	3.04	N	55
800	0.414	566.1	3.21	N	55
800	0.422	584.7	3.68	N	55
800	0.430	602.3	3.94	N	55
800	0.437	621	4.18	N	55
800	0.445	641	4.29	N	55
800	0.452	660	4.48	N	55
800	0.460	681.7	4.87	N	55
800	0.469	706.5	5.53	N	55
800	0.473	718.6	5.86	N	55
800	0.478	732	6.24	N	55
800	0.483	747.3	7.21	N	55
800	0.488	761.1	7.53	N	55
800	0.492	775.7	7.75	N	57
800	0.499	795.7	9.52	N	57
800	0.503	810.2	833	Y	57
800	0.508	827.1	847	Y	50
800	0.514	844.8	854	Y	45
800	0.519	862.4	859	Y	45
800	0.524	882	863	Y	45
800	0.535	918.6	874	Y	45
800	0.539	936	887	Y	45
800	0.558	1011.4	899	Y	45
800	0.568	1053.3	910	Y	45
800	0.575	1084.4	918	Y	45
800	0.584	1121	925	Y	45
800	0.592	1159	932	Y	40
800	0.599	1197.4	940	Y	40
800	0.608	1242.3	948	Y	40

1.30% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30

800	0.054	45.5	0	N	30
800	0.093	82.3	0.1	N	40
800	0.125	114.8	0.35	N	45
800	0.147	137.6	0.39	N	50
800	0.177	172.3	0.61	N	50
800	0.199	198.2	0.82	N	50
800	0.229	237.9	1.28	N	50
800	0.252	270.1	1.56	N	50
800	0.267	291.7	1.64	N	50
800	0.287	321.6	2.48	N	50
800	0.303	348.4	2.56	N	55
800	0.317	372.1	3.56	N	55
800	0.337	407.4	2.61	N	55
800	0.353	436.9	2.82	N	55
800	0.367	463.4	3.13	N	55
800	0.380	489.7	3.49	N	55
800	0.396	525.6	3.13	N	55
800	0.408	552.3	2.77	N	55
800	0.418	575	2.91	N	55
800	0.425	592.4	3.23	N	55
800	0.434	614.4	3.28	N	55
800	0.442	634.1	3.52	N	55
800	0.452	659.8	3.73	N	55
800	0.458	675.1	3.91	N	55
800	0.465	694.6	3.98	N	55
800	0.473	717	4.48	N	55
800	0.480	737.3	4.72	N	55
800	0.488	762.7	4.55	N	55
800	0.496	787.1	4.91	N	55
800	0.505	817.7	5.88	N	55
800	0.512	839.1	6.53	N	55
800	0.517	856.3	7.88	N	55
800	0.523	876.5	879	Y	55
800	0.529	899.2	898	Y	45
800	0.536	923.8	904	Y	45
800	0.543	949.8	910	Y	45
800	0.553	987.8	921	Y	45
800	0.562	1025.3	931	Y	45
800	0.566	1044.1	935	Y	45
800	0.574	1079.9	943	Y	45
800	0.583	1118.3	952	Y	45
800	0.592	1160.5	961	Y	45
800	0.601	1204.3	970	Y	45

1.50% NCC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.067	57.2	0.07	N	40
800	0.120	109.1	0.29	N	45
800	0.145	135.9	0.43	N	50
800	0.173	167.6	0.62	N	50
800	0.198	197.6	0.78	N	50
800	0.220	225.7	0.81	N	50
800	0.252	268.9	1.15	N	50
800	0.270	295.6	1.28	N	50
800	0.292	330.6	2.13	N	55
800	0.317	371.1	2.06	N	55
800	0.347	424.7	2.12	N	55
800	0.369	468.7	2.23	N	50
800	0.389	509.1	2.4	N	50
800	0.411	557.8	2.37	N	50
800	0.424	588.3	2.44	N	50
800	0.437	620.2	2.49	N	50
800	0.447	645.9	2.78	N	55
800	0.456	670.9	2.74	N	55
800	0.465	694.6	2.86	N	55
800	0.473	717.9	3.05	N	55
800	0.480	737.6	3.07	N	55
800	0.488	763.8	3.39	N	55
800	0.493	779.1	3.6	N	55
800	0.499	797.3	3.69	N	55
800	0.505	816	3.84	N	55
800	0.509	829.5	4.25	N	55
800	0.514	844.5	4.45	N	55
800	0.520	865.3	5.05	N	55
800	0.526	888	5.8	N	55
800	0.531	906.6	6.45	N	55
800	0.537	928.4	7.09	N	55
800	0.543	949.3	8.42	N	55
800	0.545	958.5	924	Y	55
800	0.551	982	935	Y	40
800	0.555	999.1	938	Y	40
800	0.562	1026.3	945	Y	40
800	0.568	1053.8	951	Y	40
800	0.575	1080.6	956	Y	40
800	0.584	1123.8	966	Y	40

800	0.592	1162.8	977	Y	40
800	0.601	1206.9	985	Y	40

SDS – Phase Inversion

0.05% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.024	20	0	N	50
800	0.047	39.7	0	N	50
800	0.072	61.7	0	N	50
800	0.094	82.6	0.04	N	55
800	0.114	103.3	0.04	N	55
800	0.134	124.1	0.04	N	55
800	0.153	144.8	0.04	N	55
800	0.171	164.8	0.05	N	55
800	0.187	184.2	0.1	N	55
800	0.203	203.7	0.18	N	55
800	0.219	223.7	0.26	N	55
800	0.234	244.3	0.55	N	55
800	0.248	263.7	0.73	N	55
800	0.261	283.1	0.85	N	55
800	0.274	302.3	1.47	N	55
800	0.288	324	7.33	N	55
800	0.301	343.8	10.95	N	55
800	0.313	364.2	11.45	N	55
800	0.325	385	9.54	N	55
800	0.336	404.8	6.51	N	55
800	0.347	425.4	9.09	N	55
800	0.358	445.5	14.13	N	55
800	0.368	466.2	16.95	N	55
800	0.378	486.7	16.06	N	55
800	0.388	507.5	303	Y	30
800	0.398	528.5	326	Y	30
800	0.407	548.9	339	Y	30
800	0.415	568	350	Y	30
800	0.432	608.1	370	Y	30
800	0.451	657	387	Y	30
800	0.470	710	407	Y	30
800	0.495	782.9	437	Y	30

800	0.523	877.8	472	Y	30
800	0.547	964.3	504	Y	30
800	0.564	1035.1	528	Y	30
800	0.578	1095.7	550	Y	30
800	0.593	1164.9	571	Y	30
800	0.599	1193.6	581	Y	30

0.10% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.043	36.1	0	N	35
800	0.072	62.5	0.12	N	40
800	0.092	81.3	0.41	N	48
800	0.120	109.3	1.47	N	50
800	0.134	124.1	1.62	N	50
800	0.159	151.4	1.77	N	50
800	0.186	183.2	2.41	N	50
800	0.207	208.2	2.84	N	50
800	0.225	231.9	2.96	N	50
800	0.237	248.8	3.91	N	50
800	0.262	283.5	4.21	N	50
800	0.278	308.3	247.7	Y	50
800	0.292	329.4	253	Y	30
800	0.319	374.8	312	Y	30
800	0.334	400.8	328	Y	30
800	0.362	453.4	353	Y	30
800	0.383	496.8	381	Y	30
800	0.408	551	405	Y	30
800	0.425	592	425	Y	30
800	0.447	647.8	446	Y	30
800	0.464	692.9	465	Y	30
800	0.492	776.3	501	Y	30
800	0.520	866.3	536	Y	30

0.20% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
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800	0.000	0	0	N	30
800	0.033	27.1	0	N	40
800	0.065	55.8	0.02	N	50
800	0.095	84.4	0.21	N	50
800	0.139	129.2	1.37	N	55
800	0.161	153.4	2.46	N	52
800	0.191	188.7	157.6	Y	52
800	0.213	216.6	171.6	Y	30
800	0.233	243.7	178.1	Y	30
800	0.257	276.8	180.4	Y	30
800	0.289	325.8	273.4	Y	30
800	0.323	382.4	306	Y	30
800	0.364	458.2	343	Y	30
800	0.395	522	366	Y	30
800	0.421	582	387	Y	30
800	0.447	647.8	407	Y	30
800	0.478	731.7	435	Y	30
800	0.505	817.4	464	Y	30

0.30% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.043	36.1	0.04	N	35
800	0.068	58.8	0.47	N	40
800	0.095	84.3	0.56	N	48
800	0.123	112.5	0.7	N	50
800	0.147	137.5	1.44	N	50
800	0.172	166.7	2.6	N	50
800	0.190	188	183.6	Y	50
800	0.207	209.1	208	Y	30
800	0.229	237.1	244	Y	30
800	0.260	280.8	272	Y	30
800	0.294	332.8	298	Y	30
800	0.340	411.6	349	Y	30
800	0.384	498.8	380	Y	30
800	0.428	597.8	425	Y	30
800	0.454	665	453	Y	30
800	0.482	743	483	Y	30
800	0.509	830.9	521	Y	30

0.40% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.036	30	0.03	N	35
800	0.056	47.5	0.37	N	40
800	0.074	64.2	0.44	N	48
800	0.098	86.7	0.78	N	50
800	0.118	107.2	1.18	N	50
800	0.139	129.5	1.65	N	50
800	0.163	156.2	186	Y	50
800	0.177	172.2	201.7	Y	30
800	0.206	208.1	215.8	Y	30
800	0.252	269.2	240.1	Y	30
800	0.286	319.9	338	Y	30
800	0.339	409.9	414	Y	30
800	0.361	452.6	442	Y	30
800	0.402	538.6	501	Y	30
800	0.430	603.8	544	Y	30
800	0.468	703.1	602	Y	30
800	0.491	772.3	640	Y	30
800	0.513	842.9	676	Y	30

0.50% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.039	32.5	0.02	N	35
800	0.063	54.2	0.16	N	40
800	0.089	78.3	1.27	N	48
800	0.111	99.4	1.74	N	50
800	0.135	124.8	2.85	N	50
800	0.159	151.6	179.9	Y	50
800	0.198	197.1	227.2	Y	30
800	0.243	257	287.2	Y	30
800	0.317	371.7	372	Y	30
800	0.378	485.9	451	Y	30
800	0.439	625.8	538	Y	30
800	0.464	691.8	580	Y	30

800	0.485	753.8	613	Y	30
800	0.505	815.3	646	Y	30
800	0.518	859.8	671	Y	30

Run 6 - 1.00% SDS 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.026	21.5	0	N	50
800	0.052	43.5	0.11	N	50
800	0.075	64.4	0.66	N	50
800	0.091	80.4	114.9	Y	50
800	0.099	87.7	112.8	Y	30
800	0.130	119.3	120.6	Y	30
800	0.149	139.6	186.5	Y	30
800	0.201	201	222.3	Y	30
800	0.243	256.5	306	Y	30
800	0.280	310.9	346	Y	30
800	0.312	363.33	390	Y	30
800	0.346	423.5	437	Y	30
800	0.382	494.1	492	Y	30
800	0.414	565.1	542	Y	30
800	0.430	602.8	562	Y	30
800	0.441	631	545	Y	30
800	0.462	687.4	573	Y	30
800	0.477	728.4	591	Y	30
800	0.495	784.1	621	Y	30
800	0.503	809.8	632	Y	30

OTAC – Phase Inversion

0.10% OTAC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.029	23.7	0	N	35
800	0.065	55.2	0	N	50

800	0.084	73.1	0.02	N	50
800	0.107	96.3	0.04	N	50
800	0.132	121.6	0.07	N	50
800	0.157	148.6	0.08	N	50
800	0.176	170.3	0.09	N	50
800	0.198	197.8	0.15	N	50
800	0.211	213.9	0.27	N	50
800	0.227	234.9	0.22	N	55
800	0.241	254.7	-	N	55
800	0.250	266.6	0.3	N	55
800	0.266	290.5	0.54	N	55
800	0.278	308.6	0.58	N	55
800	0.298	339	0.63	N	55
800	0.308	356.2	0.84	N	55
800	0.319	374.4	0.92	N	55
800	0.333	399	1.11	N	55
800	0.347	424.6	1.32	N	55
800	0.359	448.1	1.78	N	55
800	0.376	482.5	4.49	N	55
800	0.386	502.4	5.54	N	55
800	0.398	529.4	33.1	N	55
800	0.409	554.2	42.3	N	55
800	0.419	577.3	51.2	N	55
800	0.430	603.8	42.3	N	55
800	0.441	630.6	36.2	N	55
800	0.451	657.1	38.8	N	55
800	0.462	686.5	36.5	N	60
800	0.474	719.6	35.1	N	60
800	0.483	747	33.2	N	60
800	0.492	775.9	31.6	N	60
800	0.502	806.5	24.47	N	60
800	0.511	836.9	12.3	N	60
800	0.519	862.5	9.37	N	65
800	0.528	895.1	8.87	N	65
800	0.537	927.7	10.21	N	65
800	0.547	966.3	548	Y	40
800	0.554	993.8	572	Y	40
800	0.560	1016.8	588	Y	40
800	0.567	1048.9	607	Y	40
800	0.576	1086.2	625	Y	40
800	0.580	1106.2	640	Y	40
800	0.595	1176.2	674	Y	40
800	0.608	1242.6	704.000	Y	40

0.20% OTAC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.064	54.8	0.02	N	40
800	0.116	105.4	0.02	N	50
800	0.157	148.6	0.12	N	50
800	0.203	203.5	0.65	N	50
800	0.240	252.2	1.3	N	50
800	0.274	301.3	3.14	N	55
800	0.307	354.5	4.12	N	55
800	0.334	402	4.32	N	55
800	0.362	454.3	4.88	N	55
800	0.385	501.2	4.24	N	55
800	0.408	551.4	3.82	N	55
800	0.428	598.9	5.45	N	55
800	0.448	650.5	6.23	N	55
800	0.466	699.5	6.73	N	60
800	0.484	750.1	7.04	N	60
800	0.500	799.2	401	Y	60
800	0.516	852	430	Y	40
800	0.529	898.3	461	Y	40
800	0.544	953.4	493	Y	40
800	0.557	1007.8	523	Y	40
800	0.569	1054.2	551	Y	40
800	0.579	1098	573	Y	40
800	0.590	1149.2	597	Y	40

0.30% OTAC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.060	51	0	N	50
800	0.113	101.9	0.02	N	50
800	0.158	150	0.09	N	50
800	0.202	202.2	0.68	N	50
800	0.240	253	1.6	N	50
800	0.273	301	2.17	N	50
800	0.304	348.9	2.23	N	50

800	0.334	401.3	3.12	N	50
800	0.360	449.3	2.59	N	50
800	0.384	499.1	2.85	N	50
800	0.408	551.7	3.2	N	50
800	0.430	602.6	2.35	N	50
800	0.449	652.7	395	Y	40
800	0.467	702.3	445	Y	40
800	0.484	750	475	Y	40
800	0.499	795.7	508	Y	40
800	0.513	844.1	546	Y	40
800	0.531	904.9	582	Y	40
800	0.543	949.8	606	Y	40
800	0.556	1000.7	636	Y	40
800	0.567	1048.7	660	Y	40
800	0.581	1108.3	692	Y	40
800	0.592	1159	721	Y	40

0.40% OTAC 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.064	55.1	0	N	35
800	0.115	104.2	0.02	N	40
800	0.159	151.3	0.26	N	48
800	0.199	199	0.68	N	50
800	0.239	250.8	2.2	N	50
800	0.274	301.4	2.9	N	50
800	0.305	350.3	2.66	N	50
800	0.333	399.1	3.59	N	30
800	0.360	450.2	398	Y	30
800	0.384	499.3	450	Y	30
800	0.407	548.9	495	Y	30
800	0.430	604.2	533	Y	30
800	0.448	649.5	562	Y	30
800	0.466	699.3	596	Y	30
800	0.485	753.5	631	Y	30
800	0.500	799.7	665	Y	30
800	0.516	851.3	701	Y	30
800	0.531	904.3	738	Y	30
800	0.545	957.9	774	Y	30
800	0.556	1001.7	802	Y	30
800	0.567	1048	834	Y	30

800	0.578	1097.3	864	Y	30
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0.50% OTAC. 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.053	44.9	0	N	35
800	0.102	90.4	0.03	N	40
800	0.151	142.8	0.4	N	48
800	0.201	201	1.52	N	50
800	0.234	244.9	2.13	N	50
800	0.270	296.1	2.57	N	50
800	0.294	332.5	408	Y	30
800	0.321	377.6	452	Y	30
800	0.335	402.5	474	Y	30
800	0.360	449.2	537	Y	30
800	0.386	503.8	597	Y	30
800	0.406	547.8	641	Y	30
800	0.430	602.3	694	Y	30
800	0.446	644.5	733	Y	30
800	0.466	699.2	783	Y	30
800	0.485	754.5	829	Y	30
800	0.504	812.3	877	Y	30
800	0.516	854	910	Y	30
800	0.530	902.1	946	Y	30
800	0.544	955.4	983	Y	30
800	0.556	1003.6	1019	Y	30
800	0.567	1047.5	1048	Y	30
800	0.581	1107.9	1087	Y	30
800	0.589	1146.2	1110	Y	30
800	0.600	1201.6	1145	Y	30

Alcohol Ethoxylate – Phase Inversion

0.1% Alfonic 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
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800	0.000	0	0	N	30
800	0.027	22	0	N	30
800	0.052	43.5	0	N	30
800	0.075	64.7	0	N	30
800	0.098	87.2	0	N	30
800	0.123	111.9	0	N	30
800	0.142	132.8	0.02	N	30
800	0.162	154.7	0.08	N	30
800	0.181	176.9	0.2	N	30
800	0.199	198.5	0.59	N	30
800	0.216	220.5	0.99	N	30
800	0.233	242.4	2.27	N	30
800	0.249	264.9	2.55	N	30
800	0.265	288	3.96	N	35
800	0.280	310.9	1.91	N	35
800	0.294	333.1	2.88	N	35
800	0.308	356.5	1.97	N	40
800	0.321	378.7	2.71	N	40
800	0.333	400	0.86	N	40
800	0.346	422.6	0.94	N	40
800	0.357	444.9	1.12	N	40
800	0.368	466.8	1.14	N	40
800	0.380	490.5	1.2	N	40
800	0.392	514.8	1.14	N	40
800	0.401	535.3	1.22	N	40
800	0.410	555.3	1.3	N	40
800	0.419	576.9	1.54	N	45
800	0.428	599.3	1.08	N	45
800	0.437	621.2	1.32	N	45
800	0.446	643.2	1.56	N	45
800	0.454	665.8	1.38	N	45
800	0.462	687.5	1.61	N	45
800	0.470	709	1.85	N	45
800	0.478	732.5	1.76	N	45
800	0.485	753.3	2.15	N	45
800	0.492	776.1	1.71	N	50
800	0.499	798.2	1.73	N	50
800	0.506	820.9	1.88	N	50
800	0.513	843.7	2.01	N	50
800	0.520	867.3	2.03	N	50
800	0.527	891.4	2.27	N	50
800	0.533	914.4	2.4	N	50
800	0.539	937.1	2.74	N	50
800	0.545	959.3	2.76	N	50
800	0.551	981.5	2.86	N	50
800	0.557	1004	2.61	N	50

800	0.562	1027.1	2.92	N	50
800	0.567	1049.2	2.42	N	50
800	0.573	1071.8	2.88	N	50
800	0.578	1094.4	2.99	N	50
800	0.583	1116.3	3.16	N	50
800	0.587	1139.2	3.06	N	50
800	0.592	1160.7	357	Y	30
800	0.595	1172.9	362	Y	30
800	0.600	1200.4	372	Y	30
800	0.606	1231.8	380	Y	30
800	0.612	1264.4	389	Y	30
800	0.617	1288.6	396	Y	30
800	0.624	1326.4	406	Y	30
800	0.627	1344.1	410	Y	30
800	0.633	1381.6	421	Y	30
800	0.644	1444.5	437	Y	30
800	0.652	1501.7	449	Y	30

0.2% Alfonic 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.025	20.1	0	N	30
800	0.048	40.1	0	N	30
800	0.070	60.6	0	N	30
800	0.093	81.6	0.12	N	30
800	0.114	103.2	0.61	N	30
800	0.157	148.5	0.64	N	30
800	0.176	170.5	0.71	N	30
800	0.195	193.8	0.91	N	30
800	0.213	216.1	1.09	N	30
800	0.229	237.3	1.51	N	30
800	0.245	259.5	1.58	N	30
800	0.260	281.7	1.77	N	30
800	0.275	304.2	1.86	N	30
800	0.291	327.9	1.71	N	30
800	0.304	350.2	1.71	N	30
800	0.318	372.8	1.11	N	30
800	0.331	395.2	1.13	N	35
800	0.344	418.8	1.27	N	35
800	0.355	441.2	2.13	N	35
800	0.367	463.9	2.64	N	35

800	0.378	486.4	2.66	N	35
800	0.389	509.7	2.65	N	35
800	0.409	554.2	2.2	N	40
800	0.419	577.5	2.09	N	40
800	0.427	596.8	2.08	N	40
800	0.437	622	2.74	N	40
800	0.446	643.3	2.84	N	40
800	0.454	665.4	2.62	N	40
800	0.462	687.8	1.56	N	50
800	0.470	710	1.14	N	50
800	0.478	732.4	1.31	N	50
800	0.485	754.6	1.51	N	50
800	0.493	777.2	1.46	N	50
800	0.500	799	1.39	N	50
800	0.507	821.8	1.58	N	50
800	0.513	844.2	1.6	N	50
800	0.520	867.7	1.65	N	50
800	0.527	890.9	1.88	N	50
800	0.533	914.4	1.87	N	50
800	0.539	936.6	2.1	N	50
800	0.545	960	1.97	N	50
800	0.551	982	1.98	N	50
800	0.556	1003.8	2.46	N	50
800	0.562	1026.1	2.28	N	50
800	0.567	1048.2	2.83	N	50
800	0.572	1071.1	357	Y	30
800	0.578	1094.1	365	Y	30
800	0.583	1117.5	372	Y	30
800	0.588	1140.3	381	Y	30
800	0.593	1163.9	389	Y	30
800	0.598	1188.8	396	Y	30
800	0.602	1209.5	402	Y	30
800	0.615	1278	421	Y	30
800	0.624	1328.9	436	Y	30
800	0.636	1396.1	454	Y	30
800	0.643	1439.2	465	Y	30
800	0.653	1507	479	Y	30

0.3% Alfonic 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30

800	0.024	19.6	0	N	30
800	0.048	40.7	0	N	30
800	0.073	62.8	0	N	30
800	0.097	86.4	0	N	30
800	0.120	108.9	0	N	30
800	0.139	129.4	0.02	N	30
800	0.160	152.2	0.04	N	30
800	0.178	173.8	0.23	N	30
800	0.198	196.9	0.62	N	30
800	0.214	217.8	1.77	N	30
800	0.232	241	1.27	N	30
800	0.248	263.6	1.14	N	30
800	0.263	286.1	1.16	N	30
800	0.279	309.3	1.37	N	30
800	0.293	331.5	1.41	N	30
800	0.306	353	1.51	N	30
800	0.319	375.2	1.73	N	30
800	0.332	397.5	2.23	N	35
800	0.344	420.4	1.53	N	35
800	0.356	442.9	1.65	N	35
800	0.367	464.1	1.62	N	35
800	0.377	484.8	1.98	N	35
800	0.387	506	1.5	N	35
800	0.397	527.4	1.51	N	35
800	0.407	548.7	1.99	N	35
800	0.417	571.2	2.2	N	35
800	0.427	595.1	1.86	N	35
800	0.436	617.8	2.45	N	40
800	0.444	638.8	1.88	N	40
800	0.453	662.2	1.89	N	45
800	0.461	685.4	1.67	N	45
800	0.470	709	1.7	N	45
800	0.478	731.4	1.62	N	45
800	0.485	753.9	1.76	N	45
800	0.493	778	1.76	N	45
800	0.500	801.5	1.72	N	45
800	0.507	824.3	1.78	N	45
800	0.514	847.2	1.56	N	45
800	0.521	870.2	1.69	N	45
800	0.528	893.4	1.95	N	50
800	0.534	916.2	1.99	N	50
800	0.540	939.4	2	N	50
800	0.546	962.1	1.96	N	50
800	0.552	984.4	1.88	N	50
800	0.557	1006.3	2.015	N	50
800	0.562	1028.4	2.1	N	50

800	0.568	1050.8	355	Y	30
800	0.573	1072.6	361	Y	30
800	0.578	1093.7	369	Y	30
800	0.582	1115.3	376	Y	30
800	0.587	1136.9	383	Y	30
800	0.591	1156.6	388	Y	30
800	0.595	1177.6	394	Y	30
800	0.600	1199.1	400	Y	30
800	0.610	1251.6	415	Y	30
800	0.619	1299.9	428	Y	30
800	0.627	1345.9	441	Y	30
800	0.636	1396.5	453	Y	30
800	0.643	1437.9	463	Y	30
800	0.651	1490.8	474	Y	30
800	0.652	1501.4	478	Y	30

0.4% Alfonic 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.027	22.5	0	N	35
800	0.051	42.6	0	N	35
800	0.073	63.3	0	N	35
800	0.095	83.6	0.04	N	35
800	0.115	104.4	0.05	N	35
800	0.136	126.3	0.18	N	35
800	0.158	150.1	0.28	N	35
800	0.177	171.8	0.36	N	35
800	0.194	192.5	0.47	N	35
800	0.212	214.6	0.35	N	35
800	0.228	236.6	0.42	N	35
800	0.244	258.1	0.52	N	35
800	0.259	279.9	0.52	N	35
800	0.275	303	0.75	N	35
800	0.290	326.1	0.74	N	35
800	0.302	346.2	0.85	N	35
800	0.315	367.6	0.85	N	40
800	0.328	390.7	0.87	N	40
800	0.340	412.1	0.81	N	40
800	0.352	434.9	0.89	N	40
800	0.364	457.6	0.79	N	45
800	0.375	480.1	0.82	N	45

800	0.386	502.8	0.93	N	50
800	0.396	524.8	0.96	N	50
800	0.406	547.7	0.98	N	50
800	0.416	570.2	0.97	N	50
800	0.426	593.4	1.07	N	50
800	0.435	615.4	1.15	N	55
800	0.444	638.2	1.12	N	55
800	0.452	660.4	1.13	N	60
800	0.460	682	1.01	N	60
800	0.468	703.2	1.05	N	60
800	0.476	725.5	0.97	N	60
800	0.483	747.8	1.08	N	60
800	0.491	771.2	1.2	N	60
800	0.498	794.4	1.18	N	60
800	0.505	816.4	1.03	N	60
800	0.512	839.8	1.2	N	60
800	0.519	861.8	1.28	N	60
800	0.525	885.7	1.39	N	60
800	0.532	907.8	1.37	N	60
800	0.538	930.3	1.45	N	60
800	0.544	952.8	1.44	N	60
800	0.549	975.6	1.68	N	60
800	0.555	997.4	382	Y	60
800	0.560	1019.8	393	Y	30
800	0.566	1042.8	401	Y	30
800	0.571	1066.2	409	Y	30
800	0.576	1088.4	418	Y	30
800	0.581	1108.7	424	Y	30
800	0.585	1129.3	431	Y	30
800	0.595	1174.8	445	Y	30
800	0.605	1226.7	460	Y	30
800	0.617	1291	477	Y	30
800	0.625	1333.5	489	Y	30
800	0.632	1376.4	500	Y	30
800	0.641	1427.4	513	Y	30
800	0.652	1500.7	529	Y	30

0.5% Alfonic 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	40
800	0.013	10.5	0	N	40

800	0.027	22.6	0	N	40
800	0.049	40.9	0	N	40
800	0.071	61	0	N	40
800	0.092	81.5	0	N	50
800	0.112	101.2	0	N	50
800	0.131	120.8	0.09	N	50
800	0.151	142.3	0.13	N	50
800	0.172	165.8	0.16	N	50
800	0.189	186.7	0.26	N	50
800	0.207	208.9	0.39	N	50
800	0.223	229.7	0.64	N	50
800	0.239	250.7	0.89	N	55
800	0.254	272.2	1.05	N	55
800	0.268	292.8	1.3	N	55
800	0.282	313.6	0.9	N	55
800	0.296	335.7	0.86	N	55
800	0.309	357.1	1.03	N	55
800	0.322	380.4	1.09	N	55
800	0.334	402	1.14	N	55
800	0.346	423.7	1.42	N	55
800	0.358	446.6	1.39	N	55
800	0.370	469.2	1.85	N	55
800	0.381	492	1.72	N	55
800	0.391	513.7	1.81	N	55
800	0.401	535.8	1.97	N	55
800	0.410	556.3	2.07	N	55
800	0.419	576.8	2.61	N	55
800	0.428	598.5	2.58	N	55
800	0.437	620.2	3.2	N	55
800	0.445	640.7	3.09	N	55
800	0.452	660.2	2.94	N	55
800	0.459	679	3.32	N	55
800	0.466	697.1	3.44	N	55
800	0.473	717.4	3.31	N	55
800	0.480	738.9	3.31	N	55
800	0.487	760.1	1.17	N	55
800	0.494	781.1	6.22	N	55
800	0.501	803.8	372	Y	55
800	0.508	825.5	382	Y	40
800	0.514	846.7	391	Y	40
800	0.520	867.8	400	Y	40
800	0.527	889.6	410	Y	40
800	0.532	910.600	416	Y	40
800	0.538	933	426	Y	40
800	0.545	956.5	433	Y	40
800	0.550	977.9	441	Y	40

800	0.555	999.7	448	Y	40
800	0.561	1021.4	455	Y	40
800	0.569	1057.3	467	Y	40
800	0.580	1106.7	479	Y	40
800	0.589	1147.1	490	Y	40
800	0.598	1191.8	500	Y	40
800	0.607	1235.9	510	Y	40

Cetyl Betaine – Phase Inversion

0.1% Amphosol 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	40
800	0.027	22.5	0	N	40
800	0.053	44.7	0	N	50
800	0.076	66.2	0.06	N	50
800	0.097	86.2	0.08	N	50
800	0.119	108.4	0.08	N	50
800	0.140	130.7	0.3	N	50
800	0.160	152.4	0.41	N	50
800	0.177	172.3	0.65	N	50
800	0.195	194	0.8	N	50
800	0.211	214.5	0.74	N	50
800	0.227	234.4	0.86	N	55
800	0.243	256.5	1.01	N	55
800	0.257	277.3	1.23	N	55
800	0.272	298.7	1.58	N	55
800	0.286	321.1	2.69	N	55
800	0.300	342.7	3.51	N	55
800	0.313	365.1	4.34	N	60
800	0.325	385.7	5.17	N	55
800	0.336	405.4	6.52	N	55
800	0.348	426.9	8.88	N	55
800	0.359	448.3	10.97	N	55
800	0.370	468.9	12.11	N	55
800	0.380	490.2	22.15	N	55
800	0.391	512.6	21.29	N	55
800	0.401	534.6	16.4	N	55
800	0.410	555.3	17.46	N	60
800	0.419	576.7	17.66	N	60

800	0.428	598.1	18.52	N	60
800	0.436	619.4	19.6	N	60
800	0.445	641.5	21.53	N	60
800	0.453	662.5	21.57	N	60
800	0.461	685	20.23	N	60
800	0.469	707	539	Y	30
800	0.477	730	551	Y	30
800	0.485	752.2	564	Y	30
800	0.492	774	580	Y	30
800	0.499	796.4	591	Y	30
800	0.509	828.6	590	Y	30
800	0.519	862.2	609	Y	30
800	0.534	915.8	636	Y	30
800	0.547	964.8	661	Y	30
800	0.559	1012.9	682	Y	30
800	0.574	1079.2	711	Y	30
800	0.588	1140	732	Y	30
800	0.602	1207.6	758.000	Y	30

0.2% Amphosol 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.021	17.4	0.03	N	40
800	0.048	39.9	0.04	N	50
800	0.070	60.2	0.25	N	50
800	0.093	82.3	0.45	N	50
800	0.115	104.1	0.34	N	50
800	0.136	126	0.43	N	50
800	0.155	147.3	0.48	N	50
800	0.173	167.1	0.59	N	50
800	0.191	188.5	1.5	N	50
800	0.208	210.2	1.7	N	50
800	0.225	232.1	1.78	N	50
800	0.240	253	1.72	N	50
800	0.256	275.6	2.14	N	50
800	0.272	299.1	1.58	N	55
800	0.286	320.4	2.51	N	55
800	0.300	342.9	2.57	N	55
800	0.312	363.4	3.01	N	55
800	0.326	386.9	3.99	N	55
800	0.339	409.4	3.74	N	55

800	0.350	431.3	6.78	N	55
800	0.362	454.1	6.9	N	55
800	0.373	475.5	9.32	N	60
800	0.384	499.4	17.69	N	60
800	0.394	520.3	511	Y	40
800	0.404	542.1	539	Y	40
800	0.413	563.9	560	Y	40
800	0.423	586.1	581	Y	40
800	0.432	608.2	599	Y	40
800	0.441	630	617	Y	40
800	0.449	652.2	635	Y	40
800	0.458	675.8	652	Y	40
800	0.465	696.2	668	Y	40
800	0.473	719.2	683	Y	40
800	0.481	741.2	700	Y	40
800	0.489	764.2	715	Y	40
800	0.496	787.4	730	Y	40
800	0.509	830.7	758	Y	40
800	0.518	858.8	776	Y	40
800	0.528	893.2	796	Y	40
800	0.530	902.2	802	Y	40
800	0.543	950.8	829	Y	40
800	0.555	999.3	853	Y	40
800	0.567	1045.8	877	Y	40
800	0.577	1091.9	901	Y	40
800	0.584	1124	915	Y	40
800	0.596	1182.1	944	Y	40
800	0.605	1223	959	Y	40

0.3% Amphosol 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.024	19.4	0	N	40
800	0.046	39	0	N	50
800	0.070	60.6	0.03	N	50
800	0.093	81.8	0.28	N	50
800	0.114	102.8	0.46	N	50
800	0.135	124.8	0.77	N	50
800	0.155	146.5	0.92	N	50
800	0.173	167.1	1.02	N	50
800	0.192	190.1	1.14	N	50

800	0.209	211.8	2.34	N	50
800	0.226	233.1	2.46	N	55
800	0.242	255.8	3.77	N	55
800	0.258	278.4	3.7	N	55
800	0.274	301.5	4.9	N	55
800	0.288	322.9	6.99	N	55
800	0.301	345	5.05	N	55
800	0.315	367.9	6.01	N	55
800	0.328	391.3	8.48	N	55
800	0.340	412.8	476	Y	40
800	0.352	433.9	506	Y	40
800	0.364	457.2	533	Y	40
800	0.375	480.8	561	Y	40
800	0.387	505	587	Y	40
800	0.397	526.5	611	Y	40
800	0.407	549.3	635	Y	40
800	0.415	568.1	653	Y	40
800	0.425	590.7	675	Y	40
800	0.434	613.2	697	Y	40
800	0.443	636.9	719	Y	40
800	0.455	668	746	Y	40
800	0.463	688.6	766	Y	40
800	0.478	733.3	803	Y	40
800	0.490	767.9	832	Y	40
800	0.503	809.4	864	Y	40
800	0.516	853	896	Y	40
800	0.527	892.3	926	Y	40
800	0.539	933.5	953	Y	40
800	0.552	984.7	982	Y	40
800	0.567	1045.5	1027	Y	40
800	0.574	1076.5	1046	Y	40
800	0.587	1135	1083	Y	40
800	0.590	1150.4	1091	Y	40
800	0.601	1204.9	1123	Y	40

0.4% Amphosol 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
800	0.000	0	0	N	30
800	0.019	15.5	0	N	40
800	0.027	22.6	0	N	40
800	0.049	41.5	0	N	40

800	0.073	63.3	0.02	N	50
800	0.093	81.9	0.35	N	50
800	0.114	102.5	0.75	N	50
800	0.135	124.6	0.6	N	50
800	0.154	145.8	0.81	N	50
800	0.172	166	0.96	N	50
800	0.190	187.2	1.03	N	50
800	0.208	209.5	2.08	N	50
800	0.225	231.9	2.6	N	50
800	0.242	254.8	4.27	N	55
800	0.255	274	5.1	N	55
800	0.270	296.1	5.9	N	55
800	0.284	317.2	8.87	N	55
800	0.298	339.8	17.33	N	60
800	0.311	360.7	532	Y	30
800	0.324	383.4	573	Y	30
800	0.336	405	605	Y	30
800	0.349	428.5	638	Y	30
800	0.361	451.8	671	Y	30
800	0.372	474.1	699	Y	30
800	0.382	494.3	727	Y	30
800	0.392	515.6	750	Y	30
800	0.402	537.4	775	Y	30
800	0.415	566.6	808	Y	30
800	0.430	603.5	849	Y	30
800	0.449	652.9	898	Y	30
800	0.464	693.4	936	Y	30
800	0.480	737.4	982	Y	30
800	0.493	779	1019	Y	30
800	0.512	838	1072	Y	30
800	0.530	902.5	1129	Y	30
800	0.543	948.8	1167	Y	30
800	0.553	991.2	1202	Y	30
800	0.566	1041.9	1239	Y	30
800	0.576	1088.3	1275	Y	30
800	0.588	1142.5	1302	Y	30
800	0.596	1178	1334	Y	30
800	0.604	1217.8	1364	Y	30

0.5% Amphosol 0.01M NaCl

Oil [mL]	Actual Volume Fraction	Act. accumulative addition [mL]	Conductivity [uS/cm]	Inversion [y/n]	Mixer Setting
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800	0.000	0	0	N	40
800	0.027	22.6	0	N	40
800	0.052	44	0	N	40
800	0.077	66.3	0.07	N	40
800	0.100	89	0.25	N	40
800	0.122	110.8	0.46	N	50
800	0.143	133.2	0.77	N	50
800	0.162	154.4	0.94	N	50
800	0.180	175.7	1.22	N	50
800	0.198	197.7	1.78	N	50
800	0.214	218.1	4.01	N	50
800	0.230	239.2	7.13	N	50
800	0.246	260.7	10.2	N	50
800	0.260	281.1	15.66	N	50
800	0.275	303	465	Y	50
800	0.288	324.2	506	Y	30
800	0.302	345.8	513	Y	30
800	0.314	366.5	540	Y	30
800	0.333	399.6	589	Y	30
800	0.350	431.7	629	Y	30
800	0.371	471.3	682	Y	30
800	0.398	529.3	753	Y	30
800	0.436	617.8	856	Y	30
800	0.464	693.8	930	Y	30
800	0.503	810.8	1050	Y	30
800	0.532	908	1144	Y	30
800	0.558	1008.3	1229	Y	30
800	0.571	1066.7	1278	Y	30
800	0.587	1136.1	1333	Y	30
800	0.589	1144.5	1345	Y	30

ii. SFT/IFT

NCC – SFT/IFT

	Water	0.1%	0.2%	0.5%	1.0%	2.0%
	SFT	NCC	NCC	NCC	NCC	NCC
Drop	[mN/m]	SFT	SFT	SFT	SFT	SFT
		[mN/m]	[mN/m]	[mN/m]	[mN/m]	[mN/m]
1	63.78	65.03	60.53	64.68	63.9	64.51
2	64.41	63.92		64.84	63.35	63.01
3	64.05	63.08	63.25	64.29	63.6	60.7
4	63.71	63.47	62.24	63.81	63.43	62.79
5	62.91	63.26		64.18	62.74	61.94
6	66.13	62.63		63.79	63.23	62.09
7	63.16	63.24	60.49	64.24	63.45	61.84

8	61.78	63.15	63.37	63.97	63.43	62.03
9	64.52	63.34	61.01	63.98	64.09	61.52
10	62.23	62.92	60.38	63.91	63.34	62.02
Avg	63.67	63.40	61.61	64.17	63.46	62.25
std	1.25	0.66	1.32	0.36	0.37	1.02

[NCC]	IFT
0	57.2
0.1	42.43
0.2	38.29
0.5	52.18
1	55.02
2	48.09

SDS – SFT/IFT

	0.1% SDS	0.2% SDS	0.5% SDS	1.0% SDS
	SFT	SFT	SFT	SFT
Drop	[mN/m]	[mN/m]	[mN/m]	[mN/m]
1	40.18	33.09	33.2	32.88
2	42.13	34.03	33.3	33.63
3	41.27	38.09	33.88	34.35
4	43.79	37.57	33.26	34.52
5	45.87	39.19	33.06	35.33
6	45.78	38.94	34.35	33.37
7	47.15	40.06	33.91	33.02
8	48.44	39.12	34.12	33.32
9		39.93	33.5	33.55
10		39.8	33.66	33.4
Avg	44.33	37.98	33.62	33.74
std	2.95	2.47	0.43	0.76

[SDS]	IFT
0	57.2
0.1	4.61
0.2	5.53
0.5	7.92
1	3.02

OTAC – SFT/IFT

	0.1%	0.2%	0.5%	1.0%
	OTAC	OTAC	OTAC	OTAC
	SFT	SFT	SFT	SFT
Drop	[mN/m]	[mN/m]	[mN/m]	[mN/m]
1	55.75	48.83	40.78	38.8
2	54.65	49.48	41.14	38.63
3	53.56	47.49	41.64	39.39
4	54.03	48.27	40.86	37.33
5	55.3	47.8	39.73	37.34
6	54.99	48.66	40.37	37.38
7	56.02	49.06	40.25	37.95
8	55.16	49.02	40.97	38.55
9	53.66	49.95	40.78	38.61
10	54.88	48.82	38.94	38.22
Avg	54.80	48.74	40.55	38.22
std	0.83	0.74	0.77	0.71

[OTAC]	IFT
0	57.2
0.1	24.46
0.2	19.88
0.5	13.45
1	12.86

Alcohol Ethoxylate – SFT/IFT

	0.1% Alfonic	0.2% Alfonic	0.3% Alfonic	0.4% Alfonic	0.5% Alfonic
	SFT [mN/m]	SFT [mN/m]	SFT [mN/m]	SFT [mN/m]	SFT [mN/m]
Drop					
1	30.71	27.7	26.57	26.83	26.2
2	29.93	26.3	26.59	26.61	26.41
3	29.42	27.19	27.87	26.82	26.4
4	29.99	26.36	27.01	26.5	25.35
5	29.74	25.93	25.77	25	25.77
6	30.6	25.61	27.28	25.83	25.29
7	30.98	26.83	26.84	26.63	26.14
8	31.68	26.7	25.17	25.84	26.4
9	30.67	27.37	26.53	26.61	26.49
10	29.2	27.36	25.79	26.1	26.67
Avg	30.29	26.74	26.54	26.28	26.11
std	0.77	0.68	0.79	0.58	0.48

[Alfonic]	IFT
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0	57.2
0.1	9.68
0.2	9.9
0.3	9.91
0.4	9.19
0.5	10.1

Cetyl Betaine – SFT/IFT

	Amphosol				
	0.1%	0.2%	0.3%	0.4%	0.5%
Drop	Amphosol SFT [mN/m]	Amphosol SFT [mN/m]	Amphosol SFT [mN/m]	Amphosol SFT [mN/m]	Amphosol SFT [mN/m]
1	30.17	31.04	29.14	31.06	30.81
2	30.38	31.19	29.85	31.39	31.27
3	30.83	30.05	30.15	31.47	31.42
4	31.08	30.2	30.17	29.98	31.76
5	30.71	30.49	30.87	29.49	31.51
6	30.72	30.42	30.97	30.87	31.76
7	30.5	30.96	31.12	30.97	31.01
8	30.74	30.91	31.75	30.89	31.19
9	30.85	31.34	31.51	31.14	31.05
10	30.92	31.66	30.72	30.8	30.76
Avg	30.69	30.83	30.63	30.81	31.25
std	0.27	0.52	0.80	0.62	0.36

[Amphosol]	IFT
0	57.2
0.1	3.29
0.2	2.81
0.3	3
0.4	3.36
0.5	2.89

iii. Coalescence Stability

NCC – Coalescence Stability

Time [s]	0% NCC	0.1%NCC	0.2% NCC	0.4% NCC	0.5% NCC	0.7% NCC	0.8% NCC	0.9% NCC	1.1% NCC	1.3% NCC	1.5% NCC
0	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1	0.0918	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
2	0.1020	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
3	0.1020	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
4	0.1020	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	0.1122	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
10	0.1224	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
15	0.1327	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
20	0.1429	0.0102	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
30	0.1531	0.0143	0.0102	0.0000	0.0265	0.0306	0.0000	0.0000	0.0000	0.0000	0.0000
40	0.1571	0.0204	0.0306	0.1327	0.0816		0.1327	0.0000	0.1020	0.0000	0.1122
50	0.1735	0.0306	0.0612	0.1531	0.1327	0.1327	0.1837	0.1367	0.1571	0.1429	0.1592
60	0.1755	0.0408	0.1327	0.1735	0.1571	0.1633	0.1837	0.1673	0.1735	0.1673	0.1714
70	0.1776	0.0408	0.1633	0.1796	0.1735		0.1857	0.1857	0.1796	0.1735	0.1776
80	0.1796	0.0612	0.1735	0.1837	0.1796	0.1796	0.1878	0.1878	0.1816	0.1776	0.1796
90	0.1796	0.1122	0.1776	0.1837	0.1837	0.1857	0.1898	0.1898	0.1857	0.1816	0.1837
100	0.1816	0.1633	0.1816	0.1898	0.1878	0.1878	0.1898	0.1939	0.1878	0.1837	0.1857
110	0.1837	0.1694	0.1857	0.1898	0.1918	0.1898	0.1898	0.1878	0.1898	0.1857	0.1857
120	0.1878	0.1735	0.1878	0.1918	0.1918	0.1898	0.1939	0.2000	0.1918	0.1878	0.1878
150	0.1878	0.1837	0.1898	0.1939	0.1939	0.1918	0.1939	0.2000	0.1939	0.1898	0.1898
180	0.1878	0.1878	0.1918	0.1939	0.1939	0.1939	0.1980	0.1939	0.1980	0.1918	0.1918
240	0.1878	0.1918	0.1939	0.1959	0.1959	0.1959	0.2020	0.2000	0.1980	0.1939	0.1939
300	0.1878	0.1918	0.1939	0.1959	0.1959	0.1980	0.2020	0.2000	0.2000	0.1959	0.1939
360	0.1878	0.1918	0.1959	0.1959	0.1959	0.1980	0.2020	0.2000	0.2020	0.1980	0.1959
420	0.1898	0.1918	0.1959	0.1980	0.1959	0.2000	0.2020	0.2020	0.2020	0.2000	0.1959
480	0.1898	0.1939	0.1959	0.2000	0.1980	0.2000	0.2020	0.2020	0.2020	0.2020	0.1959
540	0.1918	0.1939	0.1959	0.2000	0.2000	0.2000	0.2020	0.2020	0.2020	0.2020	0.1980
600	0.1918	0.1939	0.1959	0.2000	0.2000	0.2000	0.2020	0.2020	0.2020	0.2020	0.1980

SDS – Coalescence Study

Time [s]	0.0% SDS	0.1% SDS	0.2% SDS	0.3% SDS	0.4% SDS	0.5% SDS	0.05% SDS
0	0.0510	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
2		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
3		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
4		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	0.0612	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
10		0.0000	0.0102	0.0102	0.0102	0.0000	0.0000
15		0.0000	0.0000	0.0000	0.0000	0.0020	0.0000
20	0.0714	0.0026	0.0163	0.0204	0.0245	0.0041	0.0000
30	0.0765	0.0000	0.0204	0.0204	0.0245	0.0061	0.0000
40	0.0816	0.0000	0.0204	0.0245	0.0306	0.0082	0.0000
50	0.0837	0.0051	0.0204	0.0245	0.0408	0.0102	0.0000
60	0.0857	0.0064	0.0245	0.0306	0.0408	0.0122	0.0000
90	0.0867	0.0128	0.0245	0.0306	0.0449	0.0128	0.0000
120	0.0898	0.0170	0.0306	0.0408	0.0469	0.0170	0.0102
150	0.0898	0.0265	0.0408	0.0490	0.0510	0.0265	
180	0.0939	0.0337	0.0531	0.0612	0.0592		0.0143
190			0.0612	0.0714	0.0612		
200			0.0714	0.0796	0.0673	0.0449	
210		0.0490	0.0776	0.0980	0.0714	0.0469	
220		0.0551	0.0918	0.1122	0.0816	0.0490	
230		0.0592	0.1020	0.1265	0.0918	0.0510	
240	0.0969	0.0673	0.1102	0.1408	0.1020	0.0531	0.0184
250		0.0694	0.1163	0.1429	0.1122	0.0551	
260		0.0735	0.1204	0.1469	0.1204	0.0571	
270	0.0969	0.0776	0.1245	0.1490	0.1306	0.0714	
280		0.0816	0.1265	0.1510	0.1347	0.0755	
290		0.0878	0.1286	0.1510	0.1388	0.0796	
300	0.0969	0.0898	0.1306	0.1510	0.1408	0.0816	0.0170
310		0.0898				0.0918	
320		0.0918				0.0980	0.0255
330	0.0969	0.0959	0.1327	0.1510	0.1429	0.1102	0.0337
360	0.0969	0.1000	0.1347	0.1551	0.1449	0.1163	0.0459
420	0.0969	0.1082	0.1408	0.1571	0.1469	0.1184	0.0561
480	0.0969	0.1082	0.1408	0.1571	0.1490	0.1224	0.0592
540	0.0969	0.1082	0.1408	0.1571	0.1510	0.1265	0.0612
600	0.0969	0.1082	0.1408	0.1571	0.1510	0.1265	0.0663

OTAC – Coalescence Study

Time [s]	0% OTAC	0.1% OTAC	0.2% OTAC	0.3% OTAC	0.4% OTAC	0.5% OTAC
0	0	0.0000	0.0000	0.0000	0.0000	0.0000
1	0.091837	0.0000	0.0000	0.0000	0.0000	0.0000
2	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
3	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
4	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
5	0.112245	0.0000	0.0000	0.0000	0.0000	0.0000
10	0.122449	0.0204	0.0245	0.0000	0.0000	0.0000
15	0.132653	0.0551	0.0408	0.0000	0.0000	0.0000
20	0.142857	0.0816	0.0816	0.0000	0.0000	0.1020
30	0.153061	0.1796	0.1796	0.0408	0.0408	0.1531
40	0.157143	0.1837	0.1837	0.1633	0.1224	0.2755
50	0.173469	0.1837	0.1898	0.1878	0.2082	0.2959
60	0.17551	0.1898	0.1939	0.1959	0.2204	0.2980
120	0.187755	0.1918	0.2000	0.2000	0.2286	0.2980
180	0.187755	0.1939	0.2020	0.2020	0.2347	0.3020
240	0.187755	0.1939	0.2020	0.2020	0.2347	0.3020
300	0.187755	0.1939	0.2020	0.2020	0.2347	0.3061
360	0.187755	0.1939	0.2020	0.2020	0.2347	0.3061
420	0.189796	0.1939	0.2020	0.2020	0.2347	0.3061
480	0.189796	0.1939	0.2020	0.2020	0.2347	0.3061
540	0.191837	0.1939	0.2020	0.2020	0.2347	0.3061
600	0.191837	0.1939	0.2020	0.2020	0.2347	0.3061

Alcohol Ethoxylate – Coalescence Study

Time [s]	0% C12-14 Alcohol Ethoxylate	0.1% C12- 14 Alcohol Ethoxylate	0.2% C12- 14 Alcohol Ethoxylate	0.3% C12- 14 Alcohol Ethoxylate	0.4% C12- 14 Alcohol Ethoxylate	0.5% C12- 14 Alcohol Ethoxylate
0	0	0.0000	0.0000	0.0000	0.0000	0.0000
1	0.09183673	0.0000	0.0000	0.0000	0.0000	0.0000
2	0.10204082	0.0000	0.0000	0.0000	0.0000	0.0000
3	0.10204082	0.0000	0.0000	0.0000	0.0000	0.0000
4	0.10204082	0.0000	0.0000	0.0000	0.0000	0.0000
5	0.1122449	0.0000	0.0000	0.0000	0.0000	0.0000
10	0.12244898	0.0000	0.0000	0.0000	0.0000	0.0000
15	0.13265306	0.0102	0.0000	0.0000	0.0000	0.0000

20	0.14285714	0.0204	0.0102	0.0102	0.0000	0.0000
30	0.15306122	0.0510	0.0306	0.0357	0.0000	0.0204
40	0.15714286	0.0816	0.0551	0.0510	0.0000	0.0367
50	0.17346939	0.1122	0.0816	0.0755	0.0204	0.0612
60	0.1755102	0.1429	0.1122	0.1020	0.0408	0.0918
120	0.1877551	0.1837	0.1857	0.1878	0.1735	0.1735
180	0.1877551	0.1878	0.1898	0.1918	0.1857	0.1837
240	0.1877551	0.1918	0.1918	0.1939	0.1878	0.1898
300	0.1877551	0.1939	0.1918	0.1939	0.1898	0.1918
360	0.1877551	0.1939	0.1939	0.1959	0.1918	0.1918
420	0.18979592	0.1939	0.1939	0.1959	0.1918	0.1939
480	0.18979592	0.1939	0.1959	0.1980	0.1939	0.1939
540	0.19183673	0.1939	0.1980	0.1980	0.1939	0.1939
600	0.19183673	0.1939	0.1980	0.1980	0.1939	0.1939

Cetyl Betaine – Coalescence Study

Time [s]	0% Cetyl Betaine	0.1% Cetyl Betaine	0.2% Cetyl Betaine	0.3% Cetyl Betaine	0.4% Cetyl Betaine	0.5% Cetyl Betaine
0	0	0.0000	0.0000	0.0000	0.0000	0.0000
1	0.091837	0.0000	0.0000	0.0000	0.0000	0.0000
2	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
3	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
4	0.102041	0.0000	0.0000	0.0000	0.0000	0.0000
5	0.112245	0.0000	0.0000	0.0000	0.0000	0.0000
10	0.122449	0.0000	0.0102	0.0102	0.0102	0.0204
15	0.132653	0.0000	0.0143	0.0102	0.0306	0.0408
20	0.142857	0.0204	0.0306	0.0306	0.0510	0.0816
30	0.153061	0.0449	0.0510	0.0408	0.0816	0.1020
40	0.157143	0.0755	0.0714	0.0714	0.1286	0.1327
50	0.173469	0.1122	0.0918	0.1122	0.1429	0.1429
60	0.17551	0.1347	0.1327	0.1429	0.1531	0.1531
120	0.187755	0.1755	0.1796	0.1837	0.1796	0.1796
180	0.187755	0.1837	0.1898	0.1939	0.1939	0.1939
240	0.187755	0.1857	0.1918	0.2020	0.1980	0.1939
300	0.187755	0.1857	0.1939	0.2041	0.2041	0.2041
360	0.187755	0.1878	0.1959	0.2041	0.2082	0.2143
420	0.189796	0.1898	0.1980	0.2041	0.2143	0.2204
480	0.189796	0.1918	0.1980	0.2041	0.2143	0.2245
540	0.191837	0.1939	0.1980	0.2041	0.2143	0.2245
600	0.191837	0.1959	0.1980	0.2041	0.2143	0.2245