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Analyzing Emulsion Dynamics via Direct Visualization and Statistical Methodologies

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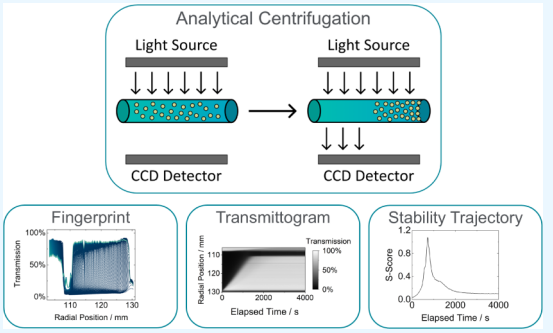
ABSTRACT: Analytical centrifugation is a powerful technique that leverages the principles of centrifugal force and optical detection to characterize emulsion droplets in a label-free and high-throughput manner. Other advantages include minimal sample preparation effort and compatibility with a wide range of emulsion formulations. However, the resulting data can be rather complex and, thus, difficult to fully understand and interpret. To tackle this, we developed two analytical methodologies that enable an easy and intuitive understanding of the data as well as an objective, quantitative analysis and validated them using six model emulsions employing different surfactants. Through their application, insights with unprecedented clarity into dynamic emulsion behavior, stability mechanisms, and emulsion-based processes can be gained, facilitating advancements in fields such as food science, pharmaceuticals, and materials engineering.



INTRODUCTION

Emulsions are ubiquitous colloidal systems found in numerous industries and natural phenomena.[1] Comprising immiscible liquid phases often stabilized by surfactants, they form stable dispersions of droplets within a continuous phase.[2] The study of emulsions is crucial due to their widespread presence and fundamental importance in various fields.[3] Furthermore, emulsions play pivotal roles in industries and consumer products due to their unique properties. In the food industry, they are vital in products like dressings, sauces, and beverages.[4,5] In pharmaceuticals and cosmetics, they serve as carriers for active ingredients.[6−8] Emulsions also find applications in agrochemicals,[9] paints and coatings,[10,11] and personal care products,[12] showcasing their versatile nature and indispensable role in manufacturing. Emulsification is also of high importance in the field of crude oil purification.[13,14]

Emulsion stability is critically important because it determines the consistency, performance, and longevity of products where emulsions are employed.[2] Stable emulsions can resist changing over time, which is essential for ensuring that their properties are preserved throughout their lifespan. Emulsions can become unstable due to a variety of reasons such as sedimentation/creaming, Ostwald ripening, and coalescence.[15−18]

Many different techniques are employed to analyze emulsions comprehensively, each offering unique insights into their properties and behavior.[19,20] For example, optical microscopy and microphotography enable direct observation of emulsion droplets, providing valuable information on size, shape, and distribution.[21−23] A common method to gauge the stability of emulsions is scanning via multiple light scattering of pulsed near-infrared light (Turbiscan).[22,24] However, as the emulsions settle in real time, measuring stable emulsions can take a long time. Dynamic light scattering (DLS) is another common method to analyze emulsions and measures fluctuations in scattered light to determine droplet size distribution but provides only a snapshot of the current state of the emulsion. Additionally, DLS struggles with polydisperse samples and lacks information about the long-term dynamic properties of emulsions. Furthermore, multiple light scattering events at high droplet concentrations can reduce the accuracy of the measurement.[3,25]

Another common tool to analyze emulsions is the analytical centrifuge. Here, in situ optical detection of droplet migration during centrifugation enables gauging droplet sedimentation velocities and size distributions in a high-throughput manner with minimal sample preparation effort and a wide range of compatible formulations. While the analytical centrifuge has also been a staple tool for emulsion analysis,[26−28] its conventional use often lacks intuitive interpretation due to the complexity of the transmission profiles. Here, we present a novel approach to provide a more intuitive understanding of

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the emulsion behavior by incorporating direct visualization of these transmission profiles. A schematic of the principles can be seen in Figure 1. In so-called transmittograms, such

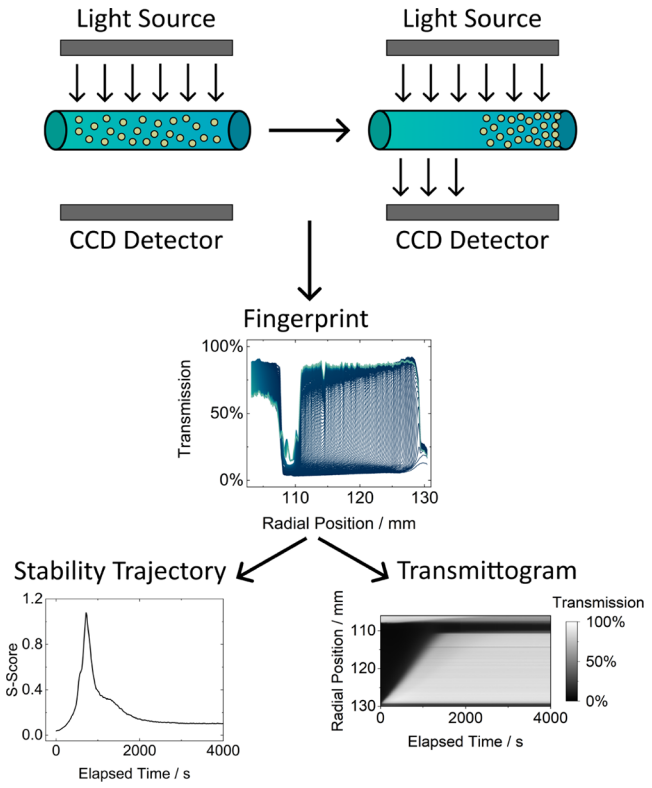

Figure 1. Measurement principle of the analytical centrifuge (top), the resulting transmission profiles of an exemplary emulsion (also known as the fingerprint, middle), and the methodologies developed to aid in analyzing and understanding the data (stability trajectory, bottom left; transmittograms, bottom right).

transmission profiles are plotted as gray values in contour plots. This methodology enables the direct observation of changes in transmission when emulsions undergo centrifugation, offering insights into stability and phase behavior as well as long-term dynamics with unprecedented clarity. In principle, this methodology can also be applied to data gathered by Turbiscan devices if the profile density is high enough (>100 profiles). Furthermore, a quantitative measure for stability is provided by evaluating S-Scores, which are z-value normalized transmission values, and their graphs as a function of the elapsed centrifugation time, called *stability trajectories*. Transmittograms and stability trajectories were originally developed by us to visualize, analyze, and optimize fuel cell catalyst ink dispersion formulations.[29,30] Within this context, studying the impact of dispersion techniques, dispersion duration, and intensity on the catalyst ink stability via these methods, and ultimately on the electrochemical performance can reveal central process−structure−property relationships.[31] This in-depth understanding enables more targeted studies and aids efforts aimed at further understanding electrochemical mechanisms and enhancing fuel cell performance. Ever since the methodology was developed, an increasing number of research groups have started to incorporate transmittograms into their dispersion analysis routine.[32,33] The versatile nature of our developed methodologies has thus encouraged us to broaden their scope to include emulsions, as well. Here, questions arise as to what extent these methodologies can be used to gauge the stability of emulsions and distinguish between different forms of instability and their sequence. As an example, sedimentation and droplet coalescence can occur simultaneously or sequentially.[18]

In this work, we focus on answering these questions using six aqueous emulsions as model systems to study and show that our methods can be readily applied to analyze emulsions. By enhancing the interpretability of analytical centrifugation data, these methodologies open up new paths for in-depth analysis and optimization of emulsion formulations across various industries.

METHODS AND MATERIALS

Materials. The emulsions were made using ultrapure Millipore water (18.2 MΩ·cm). Sodium dodecyl sulfate (>99.0%) and linalool (97.0%) were purchased from Sigma-Aldrich, didodecyldimethylammonium bromide (99.0%) was purchased from Thermofisher, and toluene (>99.9%) was purchased from VWR and used without further purification.

Emulsion Preparation. The emulsions were prepared by mixing 1 wt % of the respective surfactant (SDS, DDAB) with 89 wt % deionized water and 10 wt % of the substrate. A tip-sonicator (Branson SFX550) was used to acoustically disperse the emulsions for 5 min at an amplitude of 20 %. During sonication, the vessels containing the emulsions were submerged in ice water to prevent excessive heating. The emulsion stability was measured via an analytical centrifuge (LUMiSizer 6514−44 by LUM GmbH). For all measurements, 400 rpm (which corresponds to a relative centrifugal force of 21) and a temperature of 22 °C were used. The transmission values were measured once every 15 s until 1,000 measurements were conducted. The measurement frequency can be freely adjusted but should be chosen based on the sedimentation velocity of the droplets.

Transmittograms. As briefly discussed above, transmittograms are contour plots of the time- and space-resolved transmission data received from the analytical centrifuge after each measurement. Any program capable of creating contour plots can be used to create transmittograms from the analytical centrifugation transmission data. An Origin template that can be used to create transmittograms is available in the corresponding data publication. In a transmittogram, the *x*-axis shows the elapsed time since the beginning of the centrifugation process, and the *y*-axis shows the radial position of the measurement cell in the centrifuge. The transmission values are assigned gray tones to represent how much of the incident light was blocked through scattering phenomena at a specific time and space in the measurement cell in such a way that the darker the gray tone, the more light was blocked. The gray tone assignment can be conducted in a linear fashion or using alternative functions (e.g. logarithmic) to highlight specific areas. As time passes, one or multiple creaming/sedimentation flanks emerge that then qualitatively indicate how stable the given colloidal system is (the steeper the slope of the flanks, the less stable the system). A rising flank indicates creaming, while a falling flank indicates sedimentation, both correlating with the density difference between the organic phase and the aqueous phase.

An illustration highlighting a creaming flank can be seen in Figure 2. It also indicates how polydisperse the system is as a more polydisperse system will possess a wide range of droplet sizes that sediment/cream at different velocities, giving the

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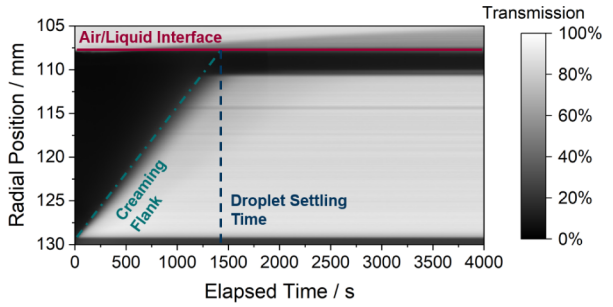

Figure 2. A transmittogram with the air/liquid interface highlighted in red, the creaming flank in green, and the time at which all droplets settle in blue.

flank a more diffuse appearance. Alternatively, a rather monodisperse system will have a sharp and well-defined sedimentation flank that readily shows the time at which all droplets have fully settled. The amount of distinct sedimentation flanks visible indicates the modality of the droplet sizes under the measuring conditions.

In summary, transmittograms show a lot of valuable and practical information about a given set of emulsion samples at once in an intuitive way. However, as a standalone visualization tool, they lack quantitative aspects that facilitate accurate and objective comparisons between samples. For this reason, the aforementioned stability trajectories were employed to supplement and complete the analysis of the emulsions via transmittograms. Another benefit of stability trajectories is that they can be calculated and interpreted fully autonomously (e.g., by a computer program) and thus open avenues for autonomous high-throughput screening and analysis of emulsions.

S-Scores and Stability Trajectories. S-Scores were developed based on statistical z-value normalization and median absolute deviation[29] and will be briefly explained in this chapter. Initially, normalized transmission profiles (Z_i) are calculated using eq 1:

$$Z_i = \frac{T_i - \overline{T_i}}{\sigma_{T_i}} \tag{1}$$

With the transmission values T_i from the analytical centrifugation data, the median transmission $\overline{T_i}$ with respect to the radial position, and the standard deviation σ_{T_i}. This transformation is used to scale the data. In the next steps, the median transmission value ($\tilde{Z}_i$, eq 2) and the median absolute deviation ($\tilde{S}_i$, the S-Score, eq 3) are calculated.

$$\tilde{Z}_i = \text{median}\,(Z_i) \tag{2}$$

$$\tilde{S}_i = \text{median}\,(|Z_i - \tilde{Z}_i|) \tag{3}$$

The S-Score then shows to what extent the transmission values are similar to each other across the measurement cell at a given time. Here, a high S-Score implies that the transmission values are dissimilar to each other with respect to the centrifugation time and the droplets are discernibly migrating, and a low S-Score means that the sample is stable. Given the nature of the median absolute deviation, the S-Score can be positive only and provides a statistically robust measure of variance.

The trace of the S-Score as a function of time is termed the stability trajectory. A rising trajectory infers how quickly the droplets migrate within the measurement cell, whereas a falling trajectory provides information about how quickly the system is approaching a clear, continuous phase. We refer to the maximum as the *bulk settling time* (BST), where most droplets will have already migrated through the sample cell. This is highlighted in Figure 3. The BST enables a quantitative

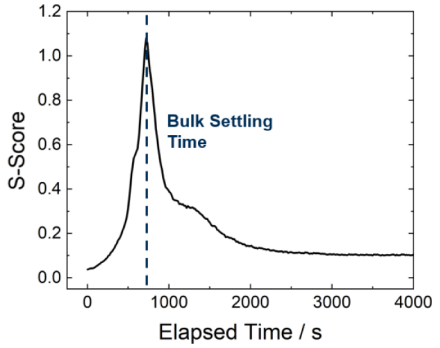

Figure 3. An exemplary stability trajectory of an emulsion. The bulk settling time is highlighted with a dashed blue line.

comparison of the emulsion stability between different samples. A mostly horizontal trajectory infers that the sample is stable and experiences no sedimentation or creaming (for example Figure 6b).

RESULTS AND DISCUSSION

To demonstrate the usefulness of the presented methodologies, six oil-in-water (O/W) emulsions were prepared that exhibit different properties. Toluene and linalool, respectively, were emulsified in a 0.1 M aqueous KOH solution using either sodium dodecyl sulfate (SDS) or didodecyldimethylammonium bromide (DDAB) as surfactants. SDS is a strongly hydrophilic anionic surfactant and is one of the most widely used emulsifiers. In contrast, DDAB is a lipophilic cationic surfactant. Two emulsions with the same oil phase but without any surfactants were prepared as references. The resulting fingerprints can be seen in Figure 4.

The emulsions without surfactant (Figure 4a, d) initially showed large gaps between each transmission line, which infers a quick settling behavior of the droplets. As one spectrum is captured every 15 seconds, a large gap between two spectra implies that a lot of droplets have migrated in this short amount of time. Emulsions with DDAB (Figure 4b, e) showed drastically improved emulsion stability. In these cases, the droplets migrated very slowly through the measurement cell. The toluene-based emulsion (Figure 4b) was slightly more stable than the linalool-based emulsion using the same surfactant (Figure 4e). This can be seen by the fraction of transmission values higher than 50% being larger in the linalool-based emulsion. The employment of SDS drastically improved the stability only for linalool-based emulsions (Figure 4f). The stability of toluene-based emulsions (Figure 4c) only moderately benefitted from the addition of SDS. For a more in-depth analysis of the emulsions, the same data set was plotted as transmittograms, which can be seen in Figure 5.

In this case, transmittograms enable an "at a glance" understanding of the emulsion dynamics and offer more insights simultaneously. Regarding the less stable emulsions without surfactant, it is now discernible that complete settling happens after around 1,000 s for the surfactant-free toluene-

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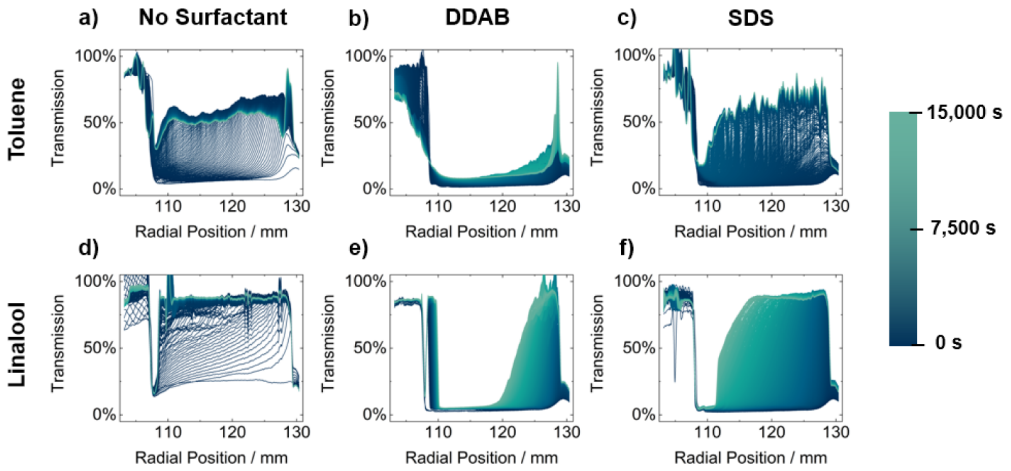

Figure 4. Transmission fingerprints of different emulsions. a), b), and c) toluene-based and d), e), f) linalool-based emulsions with three different surfactants: a) and d) no surfactant, b) and e) DDAB, c) and f) SDS. The corresponding time in the fingerprint is color-coded and can be interpreted using the legend on the right.

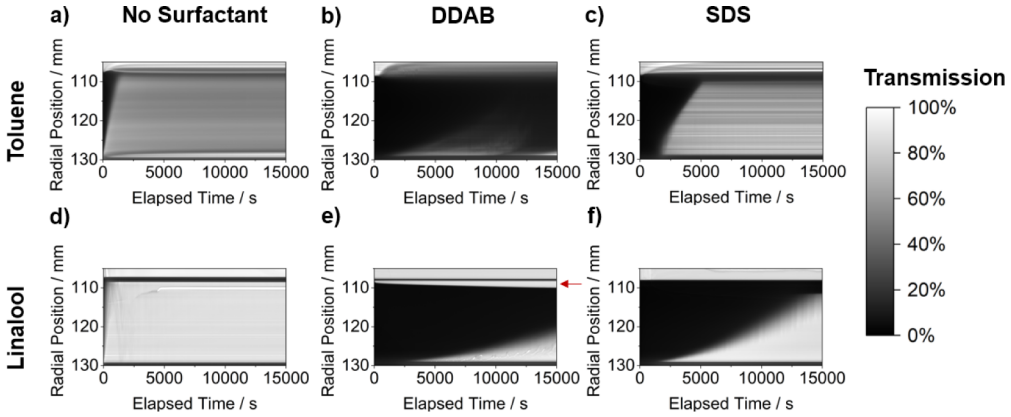

Figure 5. Transmittograms of different emulsions. a), b), and c) toluene-based and d), e), and f) linalool-based emulsions with three different surfactants: a) and d) no surfactant, b) and e) DDAB, c) and f) SDS. The grayscale legend can be found on the right. The area where rapid droplet coalescence occurs is highlighted by a red arrow in panel e).

based emulsion (Figure 5a) and around 500 s for the surfactant-free linalool-based emulsion (Figure 5d). Additionally, it is now clear that the addition of DDAB leads to creaming without droplet fusion for toluene (Figure 5b) as the transmission values remain low throughout the measurement cell. However, the addition of DDAB leads to a rapid droplet fusion for linalool (Figure 5e) as can be seen from the sharp increase in transmission values at the top of the measurement cell (highlighted by a red arrow). As separated phases are not able to scatter light like small droplets, the light passes through the measurement cell unhindered, which then leads to higher transmission values. This separated phase region is also clearly divided from the air−liquid interface by a line formed by the meniscus. Notably, the separated phase shows a slope and thus grows with time as more droplets settle. SDS-based emulsions (Figure 5c, f) did not show the same phase separation behavior and improved the overall emulsion stability when compared to the surfactant-free formulations. The creamed phase can be easily detected at the top of the measurement cell (Figure 5c) as an area with low transmission, even after the droplets have completely settled (around 5,000 s).

Similar trends are also visible in the calculated stability trajectories of the samples (Figure 6). The BSTs of the samples without surfactant (cf. Figure 6a, d and Table 1) were 465 s for toluene and 195 s for linalool. This is in line with previous conclusions on stability drawn from the transmittograms. Furthermore, the S-Score returns to a value of around 0 for linalool as the droplets fully settle. For toluene, the value stagnates at a value of around 0.25, which indicates that some fraction of the droplet remains dispersed in the continuous phase and delays the full clarification of the liquid. This is also visible in the corresponding transmittogram (Figure 5a).

Table 1. BSTs of the Analyzed Samples in Seconds

sample	BST in s
toluene, no surfactant	465
toluene, DDAB	8,040
toluene, SDS	3,180
linalool, no surfactant	195
linalool, DDAB	15,000
linalool, SDS	10,020

Comparatively, the DDAB-based aqueous emulsions showed vastly superior stability with BSTs ranging from 8,040 s for toluene and 15,000 s for linalool. Regarding toluene, the S-Score reaches a maximum value of around 0.5, indicating that no full clarification of the emulsion occurs, which confirms the corresponding transmittogram data (Figure 5b). The different

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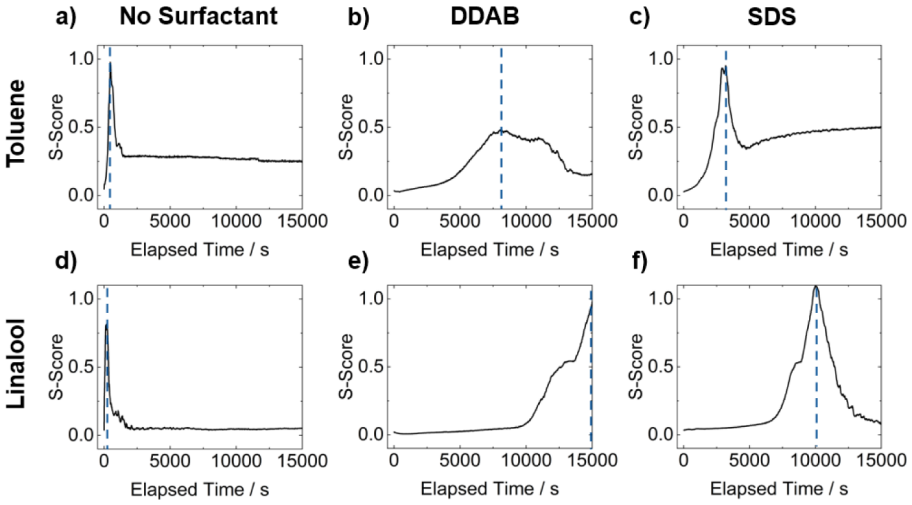

Figure 6. Stability trajectories of different emulsions. a), b), and c) toluene and d), e), and f) linalool emulsions with three different surfactants: a) and d) no surfactant, b) and e) DDAB, c) and f) SDS). The respective BST is indicated by a dashed blue line.

creaming onset times as well as the total stability of the emulsions are reflected well in the slope and the maximum of the respective stability trajectories (Figure 6b, e). The droplet coalescence, which is additionally visible in the transmittograms (Figure 5e), did not result in any significant change in the stability trajectory, highlighting the importance of using both methodologies in conjunction with each other.

SDS enhanced the stability of the emulsions as well, with BSTs ranging from 3,180 s for toluene and 10,020 s for linalool, but not as effectively as DDAB. However, with SDS-based formulations no visible droplet coalescence occurred. Overall, the BSTs correlate well with the general slope of the creaming flank and objectively gauge the stability of the emulsions. However, the minute details of the emulsion dynamics become discernible only once transmittograms are taken into account.

CONCLUSIONS

In this work, we highlight the utility of analytical centrifugation to analyze emulsions and present two analytical methodologies, transmittograms and stability trajectories, to enhance the understandability of the gathered data. These were previously developed by us with the purpose of studying the sedimentation of solids in catalyst ink dispersions but can, as demonstrated in the previous sections, also be employed to visualize and analyze the settling dynamics of emulsions as well as distinguish between different forms of emulsion instability. To this end, using toluene-based and linalool-based aqueous emulsions with different surfactants, we also showcase the ability of the analytical methodologies to readily capture the dynamic behavior of different emulsion droplet fractions and distinguish between stable and unstable, creaming, and direct droplet coalescence in emulsions. The two methodologies also allow us to quantify the stability of the formulations. For instance, the use of surfactants effectively increased the stability of aqueous linalool-based and toluene-based emulsions. The general stability of toluene-based emulsions benefited more from DDAB than from SDS. For linalool-based emulsions, DDAB produced smaller droplets initially but led to rapid droplet fusion. When using SDS, the emulsion did not show the same phase separation behavior but possessed larger droplets that migrated through the sample cell more rapidly.

The complementary nature of both methodologies is emphasized within the context of the analyzed samples. Transmittograms enable an intuitive, "at a glance" qualitative interpretation of the data, while stability trajectories give quantitative results that allow for more objective comparisons. For example, our analytical methodologies can be employed in the food industry or pharmaceuticals to gauge shelf life. In tandem, both methodologies have been shown to be effective tools to study and optimize emulsions in academia and industry alike.

ASSOCIATED CONTENT

Data Availability Statement

All data that support the findings of this work as well as an Origin template to create transmittograms is available at Zenodo (doi.org/10.5281/zenodo.13290040).

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Notes

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