

Determination of Functional Properties of Acylglycerol Emulsifier Obtained under Mild Conditions [†]

Murlykina Natalia ^{1,2,*}  and Upatova Olena ¹

¹ Department of Chemistry, Biochemistry, Microbiology and Hygiene of Nutrition, State Biotechnological University, 61051 Kharkiv, Ukraine; oiupatova@btu.kharkov.ua

² Department of Applied Chemistry, V.N. Karazin Kharkiv National University, Svobody sq., 4, 61022 Kharkiv, Ukraine

* Correspondence: nvmurlykina@karazin.ua

[†] Presented at the 4th International Electronic Conference on Applied Sciences, 27 October–10 November 2023; Available online: <https://asec2023.sciforum.net/>.

Abstract: Mono- and diacylglycerols of fatty acids are widely used as lipophilic nonionic emulsifiers and emulsion stabilizers in the production of food products. The identified deficiencies in the composition and properties of existing additives in this group created prerequisites for new developments. An acylglycerol emulsifier with essential unsaturated fatty acids based on sunflower oil was obtained under mild conditions (35–40 °C) that are harmless to human health and the environment. The evaluation of its surface-active properties was carried out using the ring tear-off and the laying drop methods. The aggregative stability of the emulsion and the number of hydrophilic-lipophilic balances were evaluated by the lifetime of individual drops of the emulsion near the interfacial surface. The results of determining the functional properties of the additive allowed us to establish its technological usage as an effective emulsifier for food systems.

Keywords: acylglycerol emulsifier; sunflower oil; the surface and interfacial tension; aggregative stability; surface interfacial tension



Citation: Natalia, M.; Olena, U. Determination of Functional Properties of Acylglycerol Emulsifier Obtained under Mild Conditions. *Eng. Proc.* **2023**, *56*, 159. <https://doi.org/10.3390/ASEC2023-15959>

Academic Editor: Nunzio Cennamo

Published: 9 November 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Lecithin, cholesterol, mono- (MAG), and diacylglycerols of fatty acids (DAG) are known natural emulsifiers [1–3]. Emulsifiers E 471, often having a hydrophilic-lipophilic balance (HLB) number of 3 to 4, are mainly composed of MAG and DAG and are often produced from palm oil [1] (pp. 79–80). They are used as lipophilic nonionic emulsifiers, emulsion stabilizers, leavening agents, texture formers, and surface-active substances for the production of margarines, table fats, broths, soups, chocolate, bakery, confectionery products and candies, drinks, ice cream and frozen desserts, food mixtures for children, etc. [2–4]. Considering the high needs of industry for lipophilic emulsifiers, it is relevant to obtain new lipophilic emulsifiers of natural origin and study their properties [4–7].

Most of the research on acylglycerol and other nonionic emulsifiers is related to the new methods of their synthesis and use [4,6,7], with the study of their antioxidant activity [5], emulsifying properties, safety, and shelf life [3–6,8]. The article [6] discusses the synthesis of acylglycerol isomers and the factors influencing acyl migration. The authors consider it important to use these acylglycerols as functional food products. The relevance of this research [7] is related to the synthesis of glycerol monolaurate and its usefulness for health. The data on the positive effect of this emulsifier on the storage and quality of food products are presented.

The formation, stability, and effectiveness of emulsions, as well as emulsion food products, depend on the functional properties of an emulsifier. The authors [9] provide a review of the physicochemical basis of the ability of mixed emulsifiers to enhance emulsion properties. Particularly, they give an overview of the most important physicochemical

properties of emulsifiers, and the origin of interactions between emulsifiers in solution and at interfaces is discussed. Studying the functional properties of emulsifiers as surface-active substances involved determining their solubility, partitioning characteristics, surface pressure, load, and activity [9]. The authors presented these data for some of the emulsifiers.

Several publications study the surface and interfacial activity of emulsifiers. However, this type of research was carried out mostly for water-soluble emulsifiers. Thus, a review [10] highlights some factors that control interfacial tension (IFT) and may have important applications in industry, for example, in the design of cosmetics, food products, and detergents.

Most of the studies have focused on oil-water systems; therefore, water-oil systems have received less attention. However, an understanding of water-oil IFT is highly desirable for the study of food systems because these systems are fundamentally different from oil-water systems due to the different chemical composition of the oil phase.

Our previous research [5,6] develops and describes a new method of obtaining the acylglycerol emulsifiers (EAGO) with nonionic surface-active substances MAG and DAG from sunflower oil under conditions of 35–40 °C. Determining the functional properties of acylglycerol emulsifiers was very important for the creation of the new food products. Therefore, we had shown the results of determining the surface and interfacial tension, the aggregative stability of the water-oil emulsion, the hydrophilic-lipophilic balance, and the emulsifying ability of the emulsifier.

2. Materials and Methods

2.1. Materials

The current study dealt with emulsifiers of acylglycerol origin (EAGO) obtained under mild conditions using laboratory equipment according to the technology of transesterification of refined sunflower oil developed by the authors [4].

A refined deodorized sunflower oil called “Oleyna Traditional” (SE Suntrade, Dnipro, Ukraine) was the main raw material used for the production of the EAGO emulsifiers. This oil is a vegetable oil of the linoleic-oleic group.

Samples 1–4 of the EAGO emulsifiers for determining the surface tension σ at the boundary with air of the water-ethanol system were made as follows: A 96% aqueous ethanol solution (Merck) was used to prepare the samples. The mass fraction of the EAGO emulsifier (W_{EAGO}) in samples of water-ethanol solutions was 0.1, 0.2, 0.3, 0.4, 0.5, and 1.0%. Samples 1–4 of the EAGO emulsifiers differed by the mass fraction of the MAG-DAG ($W_{\text{MAG-DAG}}$: 1–53.3%; 2–49.0%; 3–48.0%; 4–47.5%) and the mass ratio of $W_{\text{MAG-DAG}}/W_{\text{TAG}}$ (1–1.15; 2–0.97; 3–0.93; 4–0.91).

Highly purified paraffin FR64/66 (Am Wax, Inc., Orange, CA, USA) was used as a hydrophobic solid surface to study the interfacial interaction by the method of a lying drop.

The E471 emulsifier (S-200K, Rikevita Malaysia Sdn. Bhd., Johor Bahru, Malaysia) samples were used to compare the aggregative stability of acylglycerol emulsifiers. Solutions of the EAGO and E471 emulsifiers in oil were prepared to study the aggregative stability of the emulsion. The EAGO and E471 emulsifiers were taken for the preparation of their solutions in oil with the calculation that the mass fraction of the MAG-DAG in the oil ($W_{\text{MAG-DAG}}$) was 0.050, 0.075, 0.100, and 0.150%.

We determined the practical performance indicators of the EAGO as emulsifiers using well-known methods, which were adapted to some extent to the conditions of research on lipophilic nonionic surfactants with the use of special (HLB number) and physicochemical criteria (dispersion of heterogeneous systems, formation of hydrophobic films, stabilization of dispersed systems) [9,11–15].

2.2. Study of the Surface Tension by the Ring Tear-Off Method

The surface tension σ at the air interface of the water-ethanol system with the EAGO emulsifier samples was determined using the ring tear-off method (Du Nouy ring method) [11] using a K6 tensiometer (KRUS, Hamburg, Germany). The colloidal

characteristics of the MAG-DAG molecules of the emulsifier were calculated according to the constructed isotherm of surface tension versus concentration. The critical micelle formation concentration (CMC) was calculated. For the section of the isotherm before the CMC, we obtained the linear equation with a reliable approximation value of 0.99 and calculated the surface activity ($-d\sigma/dc$); the surface pressure of the adsorption layer of the surfactant according to the CMC (π); the limit of adsorption—Gibbs adsorption for monolayer (G_m); geometric parameters of surfactant molecules—the surface of the one surfactant molecule in a saturated adsorption layer (S) and the thickness (δ) of an adsorption layer of one molecule (axial length of a surfactant molecule), the volume of one molecule (V), adsorption work.

2.3. Study of the Interfacial Tension by the Laying Drop Method

The study of the influence of EAGO emulsifiers on interphase interaction in the process of wetting a hydrophobic surface was carried out with the method of a lying drop using laboratory equipment [11,12].

This method was based on determining the shape and size of a drop of liquid on a solid hydrophobic surface. Highly purified paraffin was used as a solid hydrophobic surface. The processes of paraffin wetting were studied at a temperature of 295 K. A drop of liquid with small dimensions (radius 0.78×10^{-6} m) and a volume of 2 μ L was applied to the surface of the paraffin using Hamilton's 10 μ L Microliter Syringe Model 701 N (Hamilton company, Reno, NV, USA). The geometric parameters of the drop were determined after its spreading and reaching the equilibrium state (exposure time was 300 s) using a micrometric eyepiece WF 10x/18 mm (Sigeta, Kyiv, Ukraine). Each measurement was repeated ten times.

Cases of liquid wetting at equilibrium (water, oil, or EAGO) on a solid hydrophobic surface of paraffin, as well as cases of variable hydrophobicity on the paraffin surface, were studied. The surface of the paraffin was changed in terms of hydrophobicity by introducing a liquid EAGO emulsifier into the paraffin with a mass fraction of 1, 3, 5, 10, 15%, or by covering the paraffin with a solid film of EAGO (100%). An increase in the mass fraction of the EAGO in paraffin made its surface more hydrophilic and significantly worsened the wetting. Such a change created favorable conditions for determining the surface tension of the liquid phases of the oil and the EAGO itself at the interface with air [13,14].

The surface tension of liquids at the interface with air σ , mN/m, was calculated based on the geometric parameters of the drop using the formula:

$$\sigma = h\rho_{\text{liq}}g(r_1^{-1} + r_2^{-1}), \quad (1)$$

where h —height of the drop, m; r_1 i r_2 —main radii of curvature, m; ρ_{liq} —liquid density; g/m³; g —acceleration of gravity; m/s².

The cosine of the equilibrium contact angle was also calculated, θ :

$$\cos\theta = (r_c^2 - h^2)/(r_c^2 + h^2), \quad (2)$$

where h —height of the drop, m; r_c —the radius of the contact area of the droplet with the surface, m.

The value of the interfacial tension of the oil and the EAGO at the interface with water were calculated based on the data of the surface tension of the liquid phases of the water, the oil, and the EAGO at the interface with air in the paraffin-liquid phase-air system, according to the thesis that the interfacial tension was represented as the difference between the surface tensions of two immiscible liquids [13,14].

2.4. Emulsion Aggregative Stability Study: The Number of Hydrophilic-Lipophilic Balances

It is known that emulsions, like all colloidal and microheterogeneous systems, are characterized by aggregative instability due to an excess of free energy at the interfacial surface. The spontaneous formation of droplet aggregates, followed by the coalescence of

individual drops with each other, is a manifestation of aggregative instability. Aggregative stability of emulsions is characterized by the speed of their separation and the time required for the coalescence (the lifetime) of individual drops under the conditions of contact with each other or with the interphase surface [9,10,15].

The aggregative stability of the emulsion stabilized by the EAGO or E 471 (S-200K) was studied based on the lifetime of its individual drops near the oil–water interface from the sides of both liquids. This research data [15] showed that a noticeable dependence of the lifetime of the drop on the size were not observed in the volume of the drop with a size of $(0.005\text{--}0.01) \times 10^{-6} \text{ m}^3$. The needed droplet size was achieved by using Hamilton's 10 μL Microliter Syringe Model 701 N (Hamilton company, Reno, NV, USA). Measurements were made using a TFA mechanical stopwatch (TFA Dostmann GmbH & Co. KG, Wertheim-Reicholzheim, Germany) on a laboratory setup according to recommendations [15]. The experiment was carried out at a temperature of 296 K and in 360 s after the formation of an interphase layer at the interface between the water and the emulsifier solution in the oil.

The type of the formed emulsion was determined by the ratio of the emulsion coalescence rate, which is inversely proportional to the droplet lifetime before the separation phase.

$$\text{oil–water emulsion formed if } \frac{v_2}{v_1} = \frac{\tau_1}{\tau_2} \geq 1, \quad (3)$$

$$\text{water–oil emulsion formed if } \frac{v_2}{v_1} = \frac{\tau_1}{\tau_2} \leq 1, \quad (4)$$

where τ_1 —the lifetime of an oil drop in water in the presence of an emulsifier; τ_2 —the lifetime of a water drop in oil in the presence of an emulsifier; v_1 —the rate of coalescence of an oil drop in water; v_2 —the rate of coalescence of a water drop in oil.

The HLB number of the EAGO and E471 was determined based on the coefficients of the emulsion coalescence rate, provided that the kinetic factors of the contact of the oil drop and the corresponding phases (water or oil) are the same [15]. The Davies formula was the basis for this:

$$\ln \frac{v_2}{v_1} = 2.2(\text{HLB} - 7). \quad (5)$$

The corresponding ratio of the times required for the coalescence of individual drops was substituted into formula (5) instead of the speed ratio, giving the final formula for calculating the HLB of the emulsifier samples:

$$\text{HLB} = 7 + (\ln \frac{\tau_1}{\tau_2})/2.2. \quad (6)$$

2.5. Statistical Analysis

For an objective judgment about the degree of confidence of the received data, a mathematical treatment of the obtained results were conducted. The reliability of the results was determined with the help of the Student's coefficients for the significance level of $p < 0.05$ and the corresponding $(n - 1)$ degrees of freedom.

In the study of surface tension by the ring tear-off method, each data point were measured at least five times to ensure reproducibility. The above mentioned procedure was repeated three times.

In the study of interfacial tension by the laying drop method and in the study of the aggregative stability of the emulsion, each data point were measured at least ten times to ensure reproducibility.

3. Results and Discussion

3.1. Study of the Surface Tension by the Ring Tear-Off Method

Like all surfactants, the MAG and DAG molecules have an amphiphilic structure. They contain polar hydrophilic—hydroxyl $-\text{OH}$; ester $-\text{OCO}-$ groups; and non-polar

hydrophobic groups of atoms—carbon chains. Due to the presence of free hydroxyl groups, MAG and DAG are more hydrophilic than triacylglycerols (TAG).

To study the dependence of the surface tension of the EAGO emulsifier with MAG-DAG on the concentration and ratio of $W_{\text{MAG-DAG}}/W_{\text{TAG}}$, the kinetic method of determining the surface tension (the ring tear-off method (Du Nouy ring method)) was applied. The interval of concentrations of the emulsifier were chosen considering the data on the content of emulsifiers in food systems. The mass fraction of the emulsifier varies in the range of 0.05–4% depending on the type of product and formulation [1–3]. The results of determining the surface tension σ are presented in Figure 1.

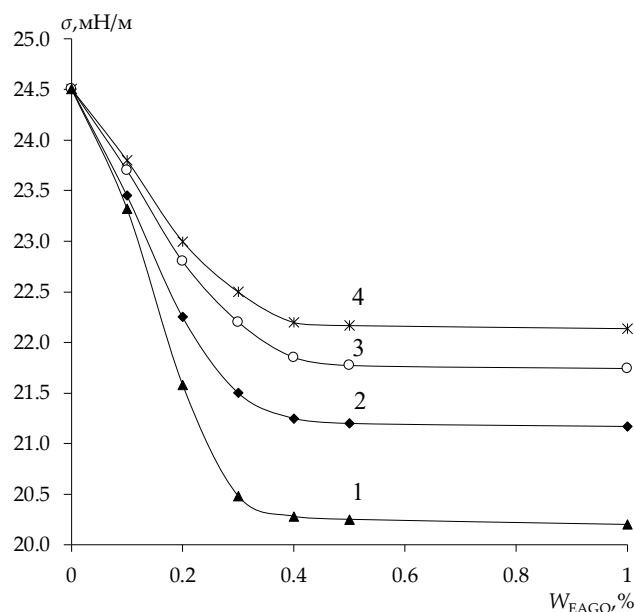


Figure 1. Dependence of the surface tension at the interface with air of the water–ethanol system on the mass fraction of the EAGO emulsifier W_{EAGO} and the $W_{\text{MAG-DAG}}/W_{\text{TAG}}$ ratio: 1–1.15; 2–0.97; 3–0.93; 4–0.91.

The plotted dependences illustrated the decrease in surface tension that is typical for surfactants. For the studied samples, this value generally decreased by 9.4–16.5% with an increase in their MAG-DAG content from 0.1 to 1.0%. That is, the surface activity of water–alcohol solutions in samples of EAGO emulsifiers at the interface liquid phase–air was insignificant, and being hydrophobic surfactants, they slightly reduced the surface tension.

The surface activity of the EAGO emulsifiers (samples 4, 3, 2, 1) in the water–ethanol–air system increased with the increase in the mass fraction of the hydrophilic MAG, DAG (4–47.5; 3–48.0; 2–49.0; 1–53.3%) and their $W_{\text{MAG-DAG}}/W_{\text{TAG}}$ ratio with hydrophobic TAG (4–0.91; 3–0.93; 2–0.97; 1–1.15). The surface tension decreased the most (from 24.51 ± 0.12 to 20.20 ± 0.14 mN/m) in the emulsifier system in sample 1.

The colloidal characteristics of the studied emulsifier samples were calculated from the surface tension isotherms (Table 1). The legends and symbols used in the table are explained in Section 2.2, “Materials and Methods”.

MAG and DAG molecules of the emulsifier sample 1 ($W_{\text{MAG-DAG}} = 53.3\%$), concentrating at the liquid–air phase separation boundary, formed the thickest adsorption layer (0.95×10^3 nm) as a result of adsorption and showed the highest surface activity ($11.60 \text{ J} \times \text{m/mol}$). The value of the calculated adsorption work of this sample was 30.99 kJ/m^2 and corresponded to the level sufficient for obtaining and stabilizing emulsions, for example, meat emulsions [2,8].

Table 1. Colloid characteristics of samples of EAGO emulsifiers.

Sample	$W_{\text{MAG-DAG}}/W_{\text{TAG}}$	CMC, mole/m ³	π , mN/m	$G_m \times 10^5$, mole/m ²	$-d\sigma/dc$, J·m/ mole	$S \times 10^{19}$, m ² /molecule	$\delta \times 10^{-3}$, nm	Adsorption Work, kJ/m ²	V , nm ³
1	1.15	3.38 ± 0.02	4.03	107.5	11.60	0.015	0.95	30.99	1.47
2	0.97	3.51 ± 0.04	3.13	72.2	3.70	0.023	0.67	30.81	1.53
3	0.93	4.35 ± 0.02	2.66	55.0	2.37	0.030	0.52	30.27	1.56
4	0.91	4.36 ± 0.03	2.31	47.6	1.87	0.035	0.45	30.26	1.58

3.2. Study of the Influence of Acylglycerol Emulsifiers on Interfacial Interaction in the Processes of Wetting a Hydrophobic Surface Using Contact Angle Measurements

The activity of oil-soluble surfactants in low-polarity environments is manifested, first of all, at the interface with water. This fact, in many cases, determines the functional surface properties of these oil-soluble surfactants.

The surface activity of the EAGO was determined by evaluating its effect on the interfacial interaction during wetting of the hydrophilized paraffin surface with water and oil. The results of research and calculations are shown in Table 2.

Table 2. The effect of the EAGO on the interfacial interaction in the paraffin–liquid phase–air system using contact angle measurements.

Surface Characteristics	Liquid Phase	Mass Fraction of EAGO in Paraffin, %						
		0	1	3	5	10	15	100
Surface tension, mN/m	water	72.35 ± 0.62	67.03 ± 0.56	62.95 ± 0.64	44.42 ± 0.70	41.53 ± 0.64	7.19 ± 0.67	4.95 ± 0.52
	oil	13.13 ± 0.46	14.73 ± 0.45	21.1 ± 0.54	20.85 ± 0.40	23.94 ± 0.38	32.91 ± 0.43	33.64 ± 0.44
	EAGO	15.11 ± 0.39	10.76 ± 0.40	12.35 ± 0.45	12.85 ± 0.50	17.45 ± 0.42	27.76 ± 0.38	29.62 ± 0.37
Cosine of the contact angle $\cos\theta_p$	water	-0.303 ± 0.004	-0.278 ± 0.005	-0.257 ± 0.009	-0.069 ± 0.001	-0.059 ± 0.001	0.886 ± 0.010	0.937 ± 0.031
	oil	0.912 ± 0.032	0.902 ± 0.029	0.895 ± 0.028	0.890 ± 0.026	0.834 ± 0.025	0.655 ± 0.020	0.635 ± 0.015
	EAGO	0.937 ± 0.035	0.933 ± 0.031	0.929 ± 0.033	0.931 ± 0.034	0.887 ± 0.026	0.819 ± 0.026	0.720 ± 0.021
Adhesion work, J/m ²	water	50.44 ± 0.05	47.80 ± 0.06	46.78 ± 0.04	41.35 ± 0.03	39.08 ± 0.04	13.56 ± 0.04	9.60 ± 0.03
	oil	25.10 ± 0.04	28.01 ± 0.05	39.99 ± 0.03	39.40 ± 0.04	43.90 ± 0.03	54.46 ± 0.05	55.01 ± 0.05
	EAGO	29.27 ± 0.04	20.80 ± 0.02	23.81 ± 0.03	24.82 ± 0.04	32.93 ± 0.03	50.48 ± 0.04	50.95 ± 0.04
Relative work of adhesion	water	0.349	0.357	0.372	0.465	0.471	0.943	0.969
	oil	0.956	0.950	0.948	0.945	0.917	0.827	0.812
	EAGO	0.968	0.967	0.964	0.966	0.944	0.909	0.860

An increase in the EAGO content in the paraffin led to an increase in the adsorption of acylglycerol molecules and their specific orientation at the paraffin–air interface. As a result, the paraffin surface was gradually hydrophilized. At the same time, the surface tension of the water decreased, and the cosine of the contact angle between the paraffin surface and the water increased. Wetting inversion was observed at 10% EAGO content. The paraffin surface became hydrophilic. At the same time, hydrophilization of the paraffin surface significantly worsened its wetting with the oil and the EAGO emulsifier and led to a decrease in the values of the cosine of the contact angle. The most significant decrease in the values of the cosines of the contact angles was observed after the introduction of 10–15% EAGO into the paraffin.

The value of the interfacial tension was calculated as the difference between the surface tension data of two immiscible liquids [11]. Therefore, the value of the surface tension of the EAGO at the water–oil interface were calculated on the basis of the obtained data on the surface tension of the water and the oil at their interface with air (liquid phase–air). The dependence of this value on the mass fraction of the EAGO in paraffin was constructed to evaluate the activity of the lipophilic EAGO at the interface between the water and the oil (Figure 2).

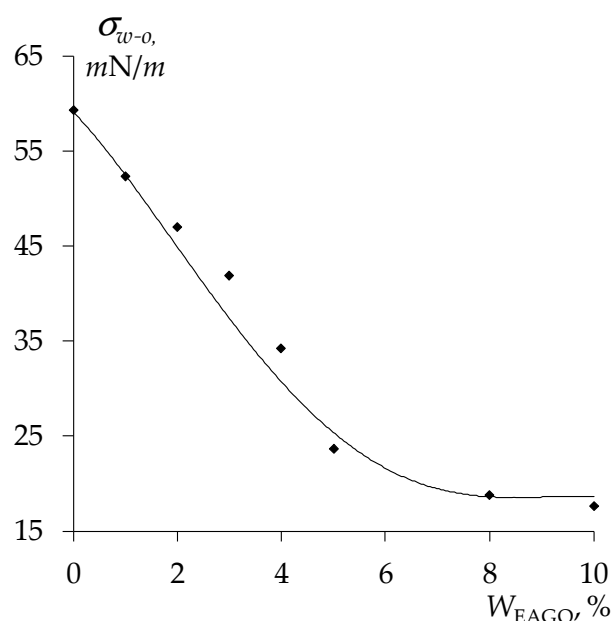


Figure 2. Dependence of the EAGO surface tension at the water-oil interface on the mass fraction of the EAGO in paraffin.

Increasing the proportion of the new additive EAGO in paraffin from 1 to 5% reduced the value of the interfacial tension at the water–oil interface from 52.3 to 23.6 mN/m, indicating sufficient surface activity.

The index of the work of adhesion is an important characteristic of the interphase interaction in heterogeneous systems. We established that in the paraffin-liquid phase-air system, the work of adhesion decreased for water and increased for oil and EAGO (Table 2), with an increase in the EAGO content in paraffin.

The data on the relative work of adhesion (Table 2) with values close to 1 indicated that the adhesion forces between the molecules of the emulsifier and the liquid phase (water or oil) in the adsorption layers on the paraffin surface approached the adhesion forces of the molecules in the volume of the emulsifier. The values of this indicator for the contacting phases of water–EAGO and oil–EAGO at the level of 0.81–0.97 indicated a high probability of the formation of a homogeneous structure in such systems in the absence of significant defects. Adsorption layers of molecules of mono- and diacylglycerols of higher carboxylic acids were formed on the hydrophilized surface of paraffin during its contact with water.

3.3. Study of the Aggregative Stability of the Emulsion: The Number of a Hydrophilic-Lipophilic Balance

Modern concepts about the mechanisms of aggregative stability of lyophobic colloidal systems differ depending on the factors regulating coagulation interactions [8–10]. Among all of the processes that reflect the instability of emulsions, only the coalescence of droplets is a characteristic irreversible process of their final destruction-separation. And the results of changes in the lifetime of the elementary emulsion droplets correlate well with the data on the strength of interfacial layers [15,16]. Therefore, studying the coalescence of emulsions is important for determining the functional properties of the EAGO emulsifier.

The HLB number is an indicator of the effectiveness of the use of emulsifiers. The HLB is expressed by the ratio of the sizes of the polar and nonpolar parts of the surfactant molecule [8,9,15,16]. HLB values vary in the range of 0 to 20. Hydrophilic emulsifiers with HLB ranging from 8 to 12 form direct oil-water emulsions. The higher the hydrophilicity, the greater the value of HLB, and the ability of surfactant molecules to form classic micelles and stabilize direct emulsions will be more pronounced. Reverse emulsions are obtained using lipophilic, oil-soluble emulsifiers with HLB values from 4 to 6. Lipophilic surfactants

with $HLB < 8$ form micellar solutions in oils. These solutions form a micellar structure with phase boundaries (micelle–medium) [9,15].

The results of studying the aggregative stability of emulsion samples stabilized by the EAGO and the E471 emulsifiers are shown in Table 3. The HLB numbers of these emulsifiers were calculated on the basis of the measured droplet lifetime near the oil–water interface from the side of both liquids (Table 3).

Table 3. Aggregative stability of the emulsion is stabilized by emulsifiers with the MAG-DAG.

Emulsifier	Mass Fraction $W_{\text{MAG-DAG}}, \%$	The Lifetime of an Oil Drop in Water τ_1, s	The Lifetime of a Water Drop in Oil τ_2, s	$\frac{\tau_1}{\tau_2} = \frac{v_1}{v_2}$	HLB
EAGO	0.050	8.8 ± 0.4	57.1 ± 0.5	0.154	6.15
	0.075	9.1 ± 0.5	59.4 ± 0.4	0.153	6.15
	0.100	9.3 ± 0.4	61.2 ± 0.5	0.152	6.15
	0.150	9.3 ± 0.3	61.6 ± 0.4	0.151	6.14
E471	0.050	2.9 ± 0.2	59.4 ± 0.5	0.049	5.63
	0.075	3.0 ± 0.3	61.4 ± 0.5	0.049	5.63
	0.100	3.1 ± 0.2	62.0 ± 0.4	0.050	5.64
	0.150	3.2 ± 0.2	62.6 ± 0.5	0.051	5.65

The results (Table 3) showed that under the conditions of obtaining an emulsion from the EAGO and E471, the stability of a water drop in the stabilized oil (τ_2) was higher than the stability of an oil drop near the interfacial surface in an aqueous solution (τ_1). This case corresponded to the formation of a water–oil emulsion. This is due to the fact that the MAG and DAG dissolve better in a dispersion medium (an oil) and are near the surface on the outside of the droplet, preventing droplet coalescence.

The stability of the emulsion stabilized by the EAGO with 0.15% MAG-DAG was at a similar level as with the E471 emulsifier. The time required for the coalescence of the drop of water in the oil near the interfacial surface was $61.6 \pm 0.4 \text{ s}$ and $62.6 \pm 0.5 \text{ s}$, respectively. The stability of the oil drop in the water near the interfacial surface was greater for the water–EAGO solution in the oil system ($9.3 \pm 0.5 \text{ s}$) than for the water–E471 solution in the oil system ($3.2 \pm 0.2 \text{ s}$). This fact can be explained by a probably more significant repulsive interaction between the particles in the interfacial adsorption layer and in the volume of the drop, probably due to the differences in the solubility of these emulsifiers and the predominance of unsaturated fatty acids in the EAGO composition [4].

The values of the HLB number of the EAGO and E471 emulsifiers were at levels of 6.1 and 5.6, respectively. The value of the HLB number of the emulsifier E471 (S-200K) correlated with the information from the manufacturer (5.3). The results obtained from this study of the aggregative stability of the emulsion with the EAGO testified about the sufficient surface strength of the adsorption layers formed by them and showed the intermolecular interaction of the MAG and DAG of the emulsifier, which can lead to a certain structuring of particles in the surface layer.

The results of the evaluation of the functional properties of the EAGO according to the system of hydrophilic–lipophilic balance and the surface and emulsifying properties proved the ability of the EAGO emulsifiers to play a significant role in the formation of stable water–oil emulsions.

4. Conclusions

The regularities of the influence of the adsorption of molecules of mono- and diacylglycerols of higher carboxylic acids on the interphase interaction of the hydrophobic surface with water and oil have been established.

Increasing the proportion of the new additive in paraffin from 1 to 5% reduced the value of the interfacial tension at the water–oil interface from 52.3 to 23.6 mN/m, which

indicated its sufficient surface activity. The decrease in surface tension in the water–oil–EAGO system and the formation of adsorption layers from the EAGO molecules at the phase separation boundary prove that it fully exhibits the functional properties of the emulsifier.

The stability of the emulsion stabilized by the new additive with 0.15% MAG-DAG was 61.6 ± 0.4 s. This indicator was 62.6 ± 0.5 s for the traditional E471 emulsifier. The HLB number of the new additive was 6.1 ± 0.1 .

The results of determining the functional properties of the additive allowed us to establish its technological usage as an effective emulsifier for food systems. It may also be promising for the creation of mixed emulsifiers.

Author Contributions: Conceptualization and methodology, M.N. and U.O.; literature analysis, M.N. and U.O.; performing experiments, M.N.; writing—original draft preparation, M.N. and U.O.; writing—review and editing, M.N. and U.O. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Akoh, C.C.; Min, D.B. *Food Lipids. Chemistry, Nutrition, and Biotechnology*, 4th ed.; Taylor & Francis Group, LLC: Boca Raton, FL, USA, 2017. [\[CrossRef\]](#)
2. Santamaria-Echart, A.; Fernandes, I.P.; Silva, S.C.; Rezende, S.C.; Colucci, G.; Dias, M.M.; Barreiro, M.F. New Trends in Natural Emulsifiers and Emulsion Technology for the Food Industry. In *Natural Food Additives*; Lage, M.Á.P., Otero, P., Eds.; Intech Open: London, UK, 2021; pp. 5–17. [\[CrossRef\]](#)
3. Nair, M.S.; Nair, D.V.T.; Johnny, A.K.; Venkitanarayanan, K. Chapter 12—Use of food preservatives and additives in meat and their detection techniques. Meat Quality Analysis. In *Advanced Evaluation Methods, Techniques, and Technologies*; Academic Press: Cambridge, MA, USA, 2020; pp. 187–213. [\[CrossRef\]](#)
4. Murlykina, N.; Upatova, O.; Yancheva, M.; Murlykina, M. Application of infrared spectroscopy for quantitative analysis of new food emulsifiers. *Ukr. Food J.* **2015**, *4*, 299–309.
5. Murlykina, N.; Upatova, O. Evaluation of the Oxidative Stability of Emulsifiers of an Acylglycerol Origin. *Biol. Life Sci. Forum* **2022**, *18*, 32. [\[CrossRef\]](#)
6. Mao, Y.; Lee, Y.-Y.; Xie, X.; Wang, Y.; Zhang, Z. Preparation, acyl migration and applications of the acylglycerols and their isomers: A review. *J. Funct. Foods* **2023**, *106*, 105616. [\[CrossRef\]](#)
7. Luo, X.; Liu, W.; Zhao, M.; Huang, Y.; Feng, F. Glycerol monolaurate beyond an emulsifier: Synthesis, in vivo fate, food quality benefits and health efficacies. *Trends Food Sci. Technol.* **2022**, *127*, 291–302. [\[CrossRef\]](#)
8. Nowicki, J. Emulsion Properties and Phase Equilibrium and of New Asymmetric Gemini Surfactants Consisting of Fatty Acid Esters of Polyethoxylated Alcohol or Phenol. *J. Surfactants Deterg.* **2010**, *13*, 195–199. [\[CrossRef\]](#)
9. McClements, D.J.; Jafari, S.M. Improving emulsion formation, stability and performance using mixed emulsifiers: A review. *Adv. Colloid Interface Sci.* **2018**, *251*, 55–79. [\[CrossRef\]](#)
10. Bui, T.; Frampton, H.; Huang, S.; Collins, I.R.; Striolo, B.A.; Michaelides, A. Water-oil interfacial tension reduction—An interfacial entropy driven process. *Phys. Chem. Chem. Rhys.* **2021**, *23*, 25075. [\[CrossRef\]](#)
11. Volpe, C.D.; Brugnara, M.; Maniglio, D.; Siboni, S.; Wangdu, T. About the possibility of experimentally measuring an equilibrium contact angle and its theoretical and practical consequences. In *Contact Angle, Wettability and Adhesion 4*; Mittal, K., Ed.; Taylor & Francis Group, LLC: Milton Park, UK; CRC Press: Boca Raton, FL, USA, 2006; pp. 79–100. [\[CrossRef\]](#)
12. Huhtamäki, T.; Tian, X.; Korhonen, J.T.; Ras, R.H.A. Surface-wetting characterization using contact-angle measurements. *Nat. Protoc.* **2018**, *13*, 1521–1538. [\[CrossRef\]](#)
13. Liu, Y.J.; Shao, J.N.; Liu, P.L. The influence of the emulsion composition on the wettability of the emulsion. *IOP Conf. Ser. Mater. Sci. Eng.* **2018**, *323*, 012013. [\[CrossRef\]](#)
14. Schmick, M.; Limage, R.; Decioyol, M.; Amtoni, M. Effect of SPAN80 on the structure of emulsified aqueous suspensions. *Colloids Surf. A Physicochem. Eng. Asp.* **2017**, *521*, 121–132. [\[CrossRef\]](#)

15. Davies, J.T. A quantitative kinetic theory of emulsion type. I. Physical chemistry of the emulsifying agent. In Proceedings of the 2nd International Congress Surface Activity, London, UK, 8–13 April 1957; pp. 426–438.
16. Pasquali, R.C.; Sacco, N.; Bregni, C. The Studies on Hydrophilic-Lipophilic Balance (HLB): Sixty Years after William C. Griffin's Pioneer Work (1949–2009). *Lat. Am. J. Pharm.* **2009**, *28*, 313–317.

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.