

Article

Techno-Economic Assessment: Food Emulsion Waste Management

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Abstract: Production of food-grade emulsions is continuously rising globally, especially in developing countries. The steepest demand growth is in the segment of inexpensive meat products where edible emulsions serve as lubricants to mitigate economic losses linked with mechanical damage during automated processing of artificial casings. Provided that production goal is to minimize emulsion transfer into the product, its vast majority becomes voluminous greasy and sticky waste. Public sewage treatment plants cannot process such waste, its cleaning processes tend to collapse under loads of emulsions. To make matters worse, composition of emulsions often changes (according to actual pricing of main components) and emulsion manufacturers carefully guard their recipes. Therefore, running of in-house sewage plants would require continuous experimentation linked with need for skilled personnel, frequent changes in technology setup and high operating costs in general. Consequently, it was repeatedly and independently reported that emulsion waste is poured onto wildlife, resulting in environmental damage and an intense rotting odor. Three new methods of emulsion breakdown are proposed and techno-economically assessed. High versatility of methods was confirmed and multiple austerity measures were incorporated. Emulsions are also assessed in terms of an energy source for aerobic and anaerobic microorganisms. It is reported that the addition of edible emulsion to compost does not result in increased product quality or cost reduction. It is firstly revealed that edible emulsions can instantly create an anaerobic environment and accelerate biogas production through the formation of surface films on feedstock surface. Adding waste food-grade emulsions to the biogas plant makes it possible to 100% reduce process water consumption in biogas stations as the process speed can be shortened by approximately 12%.

Keywords: emulsion breakdown; techno-economic assessment; bioeconomy; process management

1. Introduction

Emulsions are defined as dispersions of immiscible liquids. Many natural (milk, yolk, etc.) and processed foods (sauce, margarine, etc.) consist either partly or wholly as emulsions. Conventional

Water (W) in Oil (O) emulsion (W/O) refers to systems of microscopic spherical W droplets (usually between 100 nm and 100 μ m) dispersed in the O, whereas W/O emulsion refers to the less costly and therefore industrially favorable alternatives [1]. In addition, double and multiple emulsions, nano-emulsions (5 up to 100 nm), multilayer emulsions, colloidosomes, coated droplets, excipient emulsions, filled hydrogel particles, liposomes and micro-clusters exist [2], as it is also possible to engineer various types of, for example, W droplets dispersed within larger O droplets, which are themselves dispersed in a W system (W/O/W) [3]. These modern emulsion systems can be used to control the release of certain ingredients, reduce calories, isolate specific ingredients from each other to avoid interactions, control the release of flavor compounds, mask unpleasant flavors, etc. [4]. The process of binding feedstock into an emulsion is referred to as homogenization and it is routinely carried out using energy-demanding mechanical devices (homogenizers) to form small droplets by subjecting the feedstock to disruptive forces (high-speed blenders, high-pressure valve homogenizers, under-water macerators, colloid mills, etc.) [5]. To avoid merging of droplets upon colliding with their neighbors, gravitational separation, flocculation or coalescence stabilizers (casein, gelatin, albumin and Arabic gum, in particular) and various micro- and nano-particles are added [6,7] to improve emulsion stability (thermal and kinetic, in particular) [8]. Proteins, polysaccharides, phospholipids and saponins can be used as natural emulsifiers [9,10]. Many microorganisms produce a wide range of amphipathic compounds that exhibit surface activities [11,12]. Those that lead to the formation of stable emulsions are referred to as bioemulsifiers, such as lipopeptides, glycolipids, polysaccharides, lipopolysaccharides, proteins or lipoproteins, which are produced by a number of different bacteria (emulsan, alasan), yeasts (liposan), marine microorganisms or fungi [13,14]. Solid particles of nanoscale and microscale dimensions are also used in formation and stabilization of food emulsions (Pickering emulsions) [6,15]. The W phase may contain a variety of water-soluble ingredients, including sugars, salts, acids, bases, buffers, alcohols, surfactants, proteins, polysaccharides and preservatives [16,17]. The O phase may also contain a complex mixture of lipid-soluble components, such as triacylglycerols, diacylglycerols, monoacylglycerols, free fatty acids, sterols, vitamins, fat replacers, weighting agents, colors, flavors and preservatives [1,18]. Other ingredients are, for example, emulsifiers, thickening agents, gelling agents, weighting agents, texture modifiers, ripening inhibitors, etc. [8]. Key players in the industry quickly respond to changes in consumer preferences (higher-quality, healthier, more exotic, etc.) and produce large quantities of emulsions at minimum cost.

Nowadays, commercial emulsions are extremely resistant against any form of breakdown (storage, transport, mixing, pasteurization, sterilization, chilling, freezing, cooking, etc.), and surfactant molecules, which act as 'binding agents' and position themselves between the non-miscible elements, further enhance its stability [1,19]. Economic analysis of the emulsion industry that was recently carried out by the authors of Reference [2] indicates "huge market demand". The authors of Reference [20] point out that it is the production of affordable meat products in developing countries like India that is in critical need for emulsions and drives the global demand. This is in good agreement with Meticulous research (report ID: MRFB-104335), who expect a compound annual growth rate of 6.4% until 2025. Emulsions are needed to manage adhesion between filling and sausage tubular casing for an artificial sausage casing that is based on collagen or cellulose. However, packing of meat products into artificial casings is linked with voluminous production of emulsion waste that is of concern. There are natural mechanisms of emulsion breakdown (creaming and sedimentation, droplet aggregation, coalescence, Ostwald ripening, emulsion inversion), however, these can take decades [19,21]. The food industry necessarily needs a cheap way to manage this greasy and sticky waste of variable composition [22].

Among other things, it was also proposed to breakdown emulsions via a photo-electrochemical system for in situ generation of hydrogen peroxide [23]. The reaction is driven by photocatalytic oxidation at Pt/TiO₂ photo-anode and electro-Fenton oxidation at Fe₂O₃-loaded graphite cathode in an undivided cell. The system showed good versatility and minimal energy requirement. However, the toxic byproducts were released, and the reactant cost was rather not taken into consideration. Much effort has been put into experimentation with a high-frequency pulsed electric

field [24]. However, energy demands were high and safety concerns could be expressed. Ultrathin, nonwoven, superhydrophobic, superoleophilic fibrous poly vinylidene-fluoride membranes were also designed [25]. However, all the membranes showed insufficient capacity and limited lifetime. Numerous designs using de-emulsification enzymes of phospholipase and protease activity were patented [26]. However, these inventions can be used only towards natural emulsions obtained from aqueous extraction of lipids from plants, not against modern and strongly stabilized emulsions in the food industry [27]. Freeze/thaw de-emulsification (12.5 h at $-18\text{ }^{\circ}\text{C}$ followed by $57\text{ }^{\circ}\text{C}$) was investigated [28]. The efficiency was high; however, the process was time-consuming and energy-demanding. Some bacteria have developed the de-emulsification strategy to gain access to poorly soluble substrates and therefore, produce biosurfactants that can be characterized by the capability to breakdown various emulsions [29]. Other inventors combined biological treatment and pH adjustment [30]. However, de-emulsification capabilities of mixed bacterial population are slow, and therefore not applicable on a commercial scale [31]. Another research team presented magnetic demulsifiers with surface-active ethyl cellulose grafted on magnetic nanoparticle surfaces [32]. However, such method was shown to be environmentally and economically questionable [33]. Emulsion breakdown via micron-sized bare magnetite particles can be also traced in the literature [34]. However, the production or regeneration cost of these particles gives little hope of scale-up into the commercial dimension [35]. Repeated efforts have been made as far as de-emulsification via powerful 20 kHz ultrasound apparatus [22]. Technology showed good repeatability with lower W content (up to 40%). Their findings led them to suggest a new feasible de-emulsifying device which consists of a one-dimensional array of ultrasonic sensors which are vertically distributed to emit ultrasonic waves in the horizontal direction and in a time multiplexed manner. However, W content in up-to-date emulsions is much higher, making such a method unusable in commercial practice.

Pouring emulsions into sewer is inappropriate. Since emulsions pass almost unchanged through the grease traps [36], not only is it likely to stop the sewage treatment plant processes, to make matters worse, it also creates microscopically fine layers on the surface of any materials. As a result, any organic matter present would be protected by an extremely thin oil layer that will almost perfectly prevent it from oxidation and thus from biodegradation, which escalates the problem. Provided that gate fees (payments) for emulsion waste disposal are (as of February 2020) equal to disposal of oil and fat, and in Central Europe (Germany, Poland, Slovak, Czech Republic, Slovak Republic, Hungary, Austria, Switzerland, Slovenia) range from 150 to 200 USD t^{-1} (research conducted over the Internet at 31 waste management corporations), some “businessmen” consider “alternative” methods against which environmental reservations can be made and which are at the same time considered illegal in developed countries. A hypothesis was formed that a profitable solution can be found that could financially motivate the decision-makers to stop pouring waste emulsions out into nature. In addition to being free of hazardous chemicals and energetically undemanding, the technology should have the advantage of a high degree of versatility against various types of emulsions and being easily operated. To our knowledge, none of the published works have addressed this problem.

2. Methodology

2.1. Emulsions

2.1.1. Emulsion A

O was extracted from mechanically deshelled *Jatropha Curcas* L. seeds (acid value < 0.4) according to Reference [37]. 5 L of jatropha O was hydrogenated according to Reference [38]. Hydrogenated jatropha O was heated to $60\text{ }^{\circ}\text{C}$ and dissolved in polyglycerol polyricinoleate (E476, Spell Organics Ltd., India) using the UW200 macerator (20 min, 800 rpm, $50\text{ }^{\circ}\text{C}$) as described in Reference [39]. Subsequently, 95 L of deionized W (resistivity $> 10\text{ M}\Omega\text{cm}$, density = 0.89 gcm^{-3}) was added into the macerator (4 h, 200 rpm, $50\text{ }^{\circ}\text{C}$) to obtain 20% O/W emulsion A. Arabic gum (Esseco Srl., Italy) was added (0.5 g L^{-1}) to stabilize the emulsion.

2.1.2. Emulsion B

5 L of hydrogenated rapeseed O (acid value < 0.2) that was obtained according to Reference [40] was heated to $60\text{ }^{\circ}\text{C}$ and dissolved in polyglycerol polyricinoleate (E476, Spell Organics Ltd., India) using the UW200 macerator (20 min, 800 rpm, $50\text{ }^{\circ}\text{C}$) as described in Reference [39]. Subsequently, 95 L of deionized W (resistivity $> 10\text{ M}\Omega\text{cm}$, density = 0.91 gcm^{-3}) was added into the macerator (4 h, 200 rpm, $50\text{ }^{\circ}\text{C}$) to obtain 10% O/W emulsion B, that was stabilized via Arabic gum (1 g L^{-1}).

2.1.3. Emulsions C–I

2 (C and D) commercially available emulsions were provided from Viscofan, S.A. (Spain, Figure 1). Emulsion E was provided from Selo B.V. (The Netherlands). Devro plc (United Kingdom) provided the emulsion labeled as F, DAT-Schaub Group (Denmark) provided samples G and H. Kalle GmbH (Germany) gifted the emulsion sample labeled as I. All the commercially available emulsions are of unknown recipe.



Figure 1. Commercially available emulsions are highly stable, but the manufacturer guards and changes their production recipes rapidly (according to the price of raw materials), so it cannot be easily broken.

2.2. Emulsion Breakdown Methods

2.2.1. Method I

Sodium hydroxide (NaOH) is applied into the emulsion to increase the pH to 10. After complete dissolution and spontaneous heating of the solution, iron (III) sulfate ($\text{Fe}_2(\text{SO}_4)_3$) is added with constant and slow (5 rpm) stirring until a yellowish cloudy solution stops to settle at the bottom of the container. Subsequently, the sediment can be removed via biochar filtration.

2.2.2. Method II

Emulsion is mixed with NaOH until the pH reaches 10. Subsequently, ferric chloride solution (FeCl_3) is added slowly and with constant stirring (5 rpm) until a yellowish cloudy solution stops to settle at the bottom of the reactor. This can then be filtered off by biochar.

2.2.3. Method III

pH is increased via sodium bicarbonate (NaHCO_3) until it reaches 10. Iron (III) sulfate ($\text{Fe}_2(\text{SO}_4)_3$) is added at constant stirring (5 rpm).

2.3. Composting

Mixture (biochemical characteristics provided in Table 1, following % are given in dry matter, DM) of public greenery (grass clippings, leaves from trees and small twigs from trimming bushes, total 40%), postharvest residues (oat straw and sunflower stalks, total 40%), cattle manure (15%) and limestone (5%) was crushed using the GTS 1305G chipper (GTM, The Netherlands) to pass through a sieve with mesh of 2 cm diameter. The mixture was built into 3 m wide and 2 m high rows using the tractor-carried mobile compost turner, which was equipped with homemade apparatus to spray emulsions A and B (both 1, 5, 10 and 50 $\text{L}\cdot\text{t}^{-1}$). The composts were turned and sprayed with emulsion every 2 weeks for 4 months.

Table 1. Characteristics of the emulsion samples (A–I), where: OC = oil content; D = density; TS10 = thermal stability (at 65, 95 and 145 °C) after alkali treatment; TS3 = thermal stability (at 65, 95 and 145 °C) after acid treatment; ADS = average droplet size; BT_{DM} = biotoxicity to *Daphnia magna*; biotoxicity to *Desmodesmus subspicatus*; biotoxicity to *Sinapis alba*, p -value = 0.05.

	A	B	C	D	E	F	G	H	I
OC (%)	4.8 ± 0.7	5.0 ± 0.4	8.3 ± 1.1	8.5 ± 0.9	7.9 ± 1.3	9.2 ± 0.8	5.9 ± 1.3	4.7 ± 0.4	6.6 ± 1.0
D (g cm^{-3})	0.94 ± 0.03	0.92 ± 0.05	0.94 ± 0.02	0.93 ± 0.03	0.95 ± 0.02	0.92 ± 0.03	0.92 ± 0.01	0.96 ± 0.03	0.95 ± 0.02
TS ₆₅	yes	yes	yes	yes	yes	yes	yes	yes	yes
TS ₉₅	yes	yes	yes	yes	yes	yes	yes	yes	yes
TS ₁₄₅	yes	yes	yes	yes	yes	yes	yes	yes	yes
TS10 ₆₅	yes	yes	yes	yes	yes	yes	yes	yes	yes
TS10 ₉₅	yes	yes	yes	yes	yes	yes	yes	yes	yes
TS10 ₁₄₅	no	no	yes	yes	yes	yes	yes	yes	yes
ADS (μm)	265.2 ± 61.3	180.5 ± 72.9	104.7 ± 97.4	319.3 ± 42.7	155.7 ± 76.8	93.4 ± 63.2	145.1 ± 88.6	240.7 ± 57.4	138.3 ± 90.1
BT_{DM} (%)	100	100	100	100	100	100	100	100	100
BT_{DS} (%)	100	95	100	100	95	100	100	95	100
BT_{SA} (%)	80	95	95	85	90	100	100	85	95

2.4. Anaerobic Fermentation

Mixture (biochemical characteristics provided in Table 2, following % are given in dry matter) of maize silage (80%) and cattle manure (20%) was processed using the BRAVO 9 kW grinder (Driml s.r.o., Czech Republic) and a sieve with mesh of 2 cm diameter. Afterwards, the mixture was diluted in processing liquid (1%, 5%, 30% and 100% of emulsions and water) to obtain a mash of 12% DM. The mash was subsequently placed into series of the STIX (Stix, s.r.o., Czech Republic) semi-continuous anaerobic fermenters that were automatically operated (45 °C, 1 revolution per hour) and monitored (quantity and quality of biogas produced).

Table 2. Pricing of reactants (converted to theoretical 100% purity) in February 2020.

Reactant	Price (USD t^{-1})
sodium hydroxide	550
iron (III) sulfate	290
iron (III) chloride	610
sodium bicarbonate	190
biochar	160

2.5. Analytical Methods

An AP-5000 multimeter (AQUAREAD Ltd., United Kingdom) was used for W resistivity analysis. Acid value of O was determined via near-infrared reflectance spectroscopy (NIRS) according to Reference [41]. Soxhlet extractor (Wako Pure Chemicals, Ltd., Japan) and 99.7% hexane was used to detect level of O content (OC, %) analysis according to Reference [37]. Thermal stability at 65, 95 and 145 °C was analyzed after changing pH to 10 via sodium hydroxide (NaOH, TS10_{65, 95, 145}) or 3 via hydrochloric acid (HCl, TS3_{65, 95, 145}). Average droplet size (ADS, μm) was determined using the laser diffraction method via the Mastersizer 3000 particle size analyzer (Malvern Panalytical Ltd., United Kingdom). Sedimentation was measured by changes in height of the opaque emulsion phase in glass vials according to Reference [8]. Biototoxicity was analyzed via immobilization to crustacean *Daphnia magna* (BT_{DM}, %), acute toxicity to freshwater algae *Desmodesmus subspicatus* (BT_{DS}, %) and phytotoxicity to *Sinapis alba* (BT_{SA}, %) at 10% concentration according to Reference [42]. Cost breakdown of emulsion breakdown was carried out according to Reference [43]. Labile Pool of Carbon (LPC) was evaluated according the sulfur acid hydrolysis approach in modification by Reference [44], using the automatic, highly sensitive nitrogen and carbon analyzer (NC-90A, Shimadzu). Chemical oxygen demand (COD) and Hot-Water Extractable Nitrogen (HWEN) were analyzed as described in Reference [45]. Crude fiber (CF, sum of cellulose, hemicellulose and lignin), heavy metals (As, Cd, Cr, Cu, Hg, Mo, Ni, Pb, V and Zn), benzene, toluene and xylene (BTX), polyaromatic hydrocarbons (PAH), adsorbable organic halides (AOH), the sum of aromatic hydrocarbon compounds with 10 up to 40 carbon atoms (C₁₀–C₄₀) and polychlorinated biphenyl congeners (PCB) were detected as described by Reference [42]. Levels of furan (F) and hydroxymethyl furfural (HMF) were analyzed using the 5890 Series II gas chromatograph with a flame ionization detector (both Hewlett Packard, USA) and a DB-WAX column (Agilent technologies, USA), whereas helium was used as the carrier gas at a flow rate of 30 μLs^{-1} at a temperature of 65 °C and a nitrogen flow rate of 50 mL·min⁻¹. Compost quality was assessed by analyses on cation exchange capacity (CEC), base saturation (BS), total porosity (TP), air-filled porosity (AFP), water retention after 48 h of drainage (WR) and basal respiration (BR) according to Reference [37]. Biogas production is expressed as the number of days the substrate production reached exactly 100 L of methane (converted to 0 °C and 101.3 kPa) per 1 kg dry weight (DW). All trials were carried out with 6 repetitions.

3. Results and Discussion

A large number of compounds that show properties similar to edible emulsions randomly enter the environment, either as natural products originating from animals, plants or microorganisms, such as steroids, terpenes or waxes. However, these are not artificially stabilized and thus do not cause environmental problems [29]. Characteristics of the emulsions that were produced for research purposes “in-house” as well as characteristics of commercially available samples are stated in Table 1. Soxhlet analyses on O content and indirectly also the analyses on density confirmed that all of the samples refer to O/W emulsions. Majority of the samples were shown to have an oil concentration between 5% and 9%, which is in line with commercial practice [1]. One could argue that some production savings regarding the used oil could be realized if the oil level was lower less than 5%, which is technically possible [2]. However, respecting such a requirement, the energy needed to run (longer or intensively) the apparatuses that cause the disruptive forces sharply increases and results in an increase of total production costs. To make matters worse, the emulsions of OC lower than 5% tend to suffer from lower stability [9], which requires additional cost linked with its stabilization techniques. All emulsions showed resistance to thermal breakdown after acid treatment. Only the “homemade” emulsions showed some weaker stability after alkali treatment at the highest temperatures. In response to analyses on ADS, it can be argued that all the commercial emulsions show excellent thermal stability in general. All emulsions immobilized 100% of *Daphnia magna* crustacean and killed between 95% and 100% of *Desmodesmus subspicatus* freshwater algae, even at 10% concentration, which can be altogether interpreted as a danger to water systems [42]. Phytotoxicity to germination of *Sinapis alba*

showed slight differences (between 85% and 100%). Nevertheless, the values obtained are (in agreement with Reference [13]) sufficient to claim that all food-grade emulsions represent a risk to plants [30], respectively to soil biota [36]. With reference to actual pricing of reactants (Table 2), the effort to breakdown the emulsion samples (A–I) up to 99% efficiency showed (Table 3) that it can be stated that this goal was the most expensive (average 29.5 and median 34.8 USD·t⁻¹). Data reveals that the breakdown of the “homemade” emulsions (A and B) was one of the less expensive ones (approximately 3.5 times cheaper than others). Further analysis reveals that on average, the breakdown method I appears to be the most expensive, whereas III was the cheapest. Note that for some emulsions (E and H), the opposite is true, but it can be concluded that method III seems to be the most universal. If the ambition to breakdown the emulsions is reduced to 98%, the cost is significantly (by more than 1/5 in comparison to 99% breakdown efficiency) reduced in all cases (average 23.5 and median 23 USD·t⁻¹). Given that the requirements on emulsion breakdown efficiency are reduced to 95%, there is another significant (by more than 1/2 in comparison to 99% breakdown efficiency) reduction in cost (average 15.6 and median 14.8 USD·t⁻¹).

Table 3. Characteristics of feedstock used for anaerobic fermentation and composting (converted to DM), where: I = public greenery, II = postharvest residues, III = cattle manure and IV = maize silage, *p*-value = 0.05.

	I	II	III	IV
DW (g kg ⁻¹)	439.7 ± 75.4	613.0 ± 47.4	214.7 ± 41.4	297.1 ± 25.8
CF (g kg ⁻¹)	378.1 ± 40.3	576.6 ± 51.1	77.5 ± 30.8	225.6 ± 33.0
LPC (g kg ⁻¹)	196.3 ± 21.5	118.3 ± 18.4	31.4 ± 12.9	135.7 ± 20.2
HWEN (g kg ⁻¹)	12.0 ± 5.2	14.1 ± 7.7	17.5 ± 8.1	13.8 ± 5.1
COD (g kg ⁻¹)	816.2 ± 134.1	784.2 ± 63.2	1107.9 ± 114.7	1304.3 ± 163.5
As (mg kg ⁻¹)	1.0 ± 1.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Cd (mg kg ⁻¹)	1.0 ± 1.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Cr (mg kg ⁻¹)	33.0 ± 2.0	10.0 ± 3.0	14.0 ± 2.0	15.0 ± 2.0
Cu (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Hg (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Mo (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Ni (mg kg ⁻¹)	16.0 ± 4.0	3.0 ± 1.0	3.0 ± 2.0	4.0 ± 1.0
Pb (mg kg ⁻¹)	45.0 ± 7.0	4.0 ± 2.0	4.0 ± 3.0	5.0 ± 3.0
V (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Zn (mg kg ⁻¹)	120.0 ± 19.0	23.0 ± 9.0	15.0 ± 9.0	14.0 ± 6.0
BTX (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
PAH (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
AOH (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
C ₁₀ –C ₄₀ (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
PCB (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
F (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
HMF (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0

When the emulsion breakdown is achieved, there are two main ways to proceed. At first, it is possible to pour the broken emulsion into the sewer. Unless the volumes of oil are enormous (more than 2 k kgday⁻¹, which is equal to some 20 t of waste emulsion day⁻¹ before breakdown), a conventional sewage plant, which is designed for a city of over 10,000 residents, can easily handle such a load [46]. If such a solution is chosen and the cheapest and most universal method, III, is applied for its implementation with 99% breakage requirement, the difference between the cost of landfilling and the cost of breaking down the emulsion represents savings of some 120 USD t⁻¹ (= 150 – 29.5). However, this is a strong argument only in countries where the appropriate legislation is respected. The second alternative is to further utilize the products of emulsion breakdown. Edible oil could be used, for example, to produce low-quality soaps. Nevertheless, this does not improve the economics of

the process because the oil levels are low (50 L t^{-1}), and therefore the price (2.4 USD L^{-1}) would not be competitive.

Regarding composting efforts, all the feedstock used showed (Table 4) characteristics not far from other reports [39]. The presence of possible inhibitors in the feedstock was negligible [42], which allows a good degree of generalization. Interpreting the quality of composts treated with emulsions A (Table 5) and B (Table 6) is straightforward. There was no improvement in any of the monitored compost quality indicators in any of the cases. Both emulsions only worsened the quality of the compost produced, with the higher doses leading to greater degradation of quality. To make matters worse, all the composts smelled badly, and mold deposits often appeared (proportionally to the increased emulsion dose). In agreement with Reference [22], these observations indicate that emulsions formed thin layers on the feedstock surface, which prevented air from entering the internal structures and pores. Characteristics of blank sample was as follows: $\text{CEC} = 37.16 \pm 5.69 \text{ cmol}^+ \text{ kg}^{-1}$, $\text{BS} = 83\%$, $\text{TP} = 15\%$, $\text{AFP} = 7\%$, $\text{WR} 9\%$, $\text{BR} = 0.035 \text{ mg C-CO}_2 \text{ kg}^{-1} \text{ h}^{-1} (\%)$. It can be assumed that in particular, the decrease in basal respiration (Tables 5 and 6) is linked with reduced population dynamics of aerobic microorganisms. It seems that treatment with emulsions are locally capable of switching the aerobic composting process to anaerobic despite intensive aeration via the tractor-carried mobile compost turner.

Table 4. Cost (USD as of February 2020) needed for 99%, 98% and 95% breakdown of 1 k kg of emulsion samples (A–I) according to methods I–III.

	A	B	C	D	E	F	G	H	I
99% breakdown									
I.	29.7	27.7	41.5	66.1	19.3	34.8	26.0	36.7	35.7
II.	24.0	29.2	49.6	50.4	14.7	27.6	39.4	11.8	44.3
III.	8.5	8.2	18.5	24.7	25.0	22.3	27.9	24.9	27.8
98% breakdown									
I.	25.4	23.3	36.4	49.1	15.2	27.5	23.6	31.4	30.3
II.	22.3	26.4	34.5	44.3	9.4	22.7	31.3	8.4	35.2
III.	7.6	7.7	14.0	17.4	19.3	16.8	14.4	16.7	23.0
95% breakdown									
I.	18.0	14.5	22.1	24.7	10.3	15.8	15.6	19.9	21.5
II.	14.6	20.3	19.7	24.8	7.9	17.6	21.4	7.5	27.6
III.	7.2	7.1	9.3	11.5	13.6	13.0	11.7	12.9	14.8

Table 5. Changes in composts characteristics following treatment with emulsion A, where: V = 1 Lt^{-1} , VI = 5 Lt^{-1} , VII = 10 Lt^{-1} and VIII = 50 Lt^{-1} spraying intensity. Results are expressed as % change in comparison to blank sample (no emulsion treatment = 100%).

	V	VI	VII	VIII
CEC (%)	0	0	−4	−4
BS (%)	0	−1	−5	−10
TP (%)	0	0	−3	−5
AFP (%)	0	0	−1	−4
WR (%)	−1	−1	−3	−9
BR (%)	−2	−5	−16	−23

Table 6. Changes in composts characteristics following treatment with emulsion B, where: IX = 1 Lt⁻¹, X = 5 Lt⁻¹, XI = 10 Lt⁻¹ and XII = 50 Lt⁻¹ spraying intensity. Results are expressed as % change in comparison to blank sample (no emulsion treatment = 100%).

	IX	X	XI	XII
CEC (%)	0	0	-1	-3
BS (%)	0	0	-4	-12
TP (%)	0	0	-3	-5
AFP (%)	0	0	-2	-5
WR (%)	0	-1	-7	-16
BR (%)	-1	-2	-15	-30

With regards to anaerobic fermentation, biochemical characteristics of the feedstock (Table 3) showed normal values with no signs of inhibition [42]. All biogas experiments were quite consistent on methane concentration (45.71% ± 3.02%), whereas the blank sample produced 100 L of methane in 415 h. Biogas production always starts with a lag phase, which takes usually 2 days. During this initial period, there is almost zero biogas production and the anaerobic environment is being formed. Results on methanogenesis dynamics (Table 7) indicate that the same mechanisms occurred (regardless of emulsion A or B) as in the case of composting. It can be assumed that the thin emulsion films covered the surface of the feedstock being processed and thus accelerated the formation of the anaerobic environment. Even the feedstock that floated to the surface of the process liquid was thus in an anaerobic environment. Similar mechanisms were observed by researchers who investigated possibilities of biogas production from palm oil mill effluent [47] and those who optimized biogas production from olive oil mill wastewater [48]. It was also reported that the anaerobic process was inhibited when oil levels rose over 40% [48]. Nevertheless, such an effect did not occur when O/W emulsions were used (Table 7), as such high oil concentration could never have occurred. The data obtained do not allow to predict at what emulsion level the maximum biogas yield occurs. However, the possibility of completely replacing water by O/W emulsions is both an environmentally and economically significant finding. Despite the fact that oils are components of biomass with long decomposition time, the presented analyses all together indicate relatively good degradability. It is assumed that formation of an anaerobic film on the substrate particles is an important, but not the only, factor. It is expected (but not confirmed yet) that the presented results differ from the slowing effect most often reported in the literature because of large reaction surfaces. Plethora of microscopic droplets are highly likely to be more easily biodegradable than large fragments of oil. Almost no oil could be traced in fermentation residues (Table 8). This demonstrates that the edible oil was fully used as an immediate source of energy for consortia of anaerobic microorganisms.

Table 7. Dynamics of methane production, where: XIII = 1%, XIV = 5%, XV = 30% and XVI = 100% emulsion level. Results are expressed as % change in comparison to blank sample (water only = 100%)/hours.

	XIII	XIV	XV	XVI
A (%)	-3/403	-11/369	-11/369	-12/365
B (%)	-3/402	-10/373	-12/365	-12/365

Table 8. Oil residues in fermentation residues, where: XIII = 1%, XIV = 5%, XV = 30% and XVI = 100% emulsion level, *p*-value = 0.05.

	XIII	XIV	XV	XVI
A _{oil residue} (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.3 ± 0.2
B _{oil residue} (mg kg ⁻¹)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.1 ± 0.1

4. Conclusions

Increasing the pH to 10 via sodium bicarbonate that is followed by iron (III) sulfate precipitation and biochar filtration was shown to be the optimal edible emulsion breakdown method in terms of minimum cost and maximum versatility. However, waste management of W/O emulsions can also be environmentally friendly while generating profits. Despite the fact that oils are biomass components with long decomposition time, its emulsification significantly increases its biodegradability. It was firstly reported that after being added to the biogas plant, the following synergies can be observed. At first, it is possible to replace all process water, which means tremendous savings. Secondly, the microscopic oil droplets serve as a disposable energy source for consortia of anaerobic microorganisms and thus shorten the initial phase lag. Thirdly, the emulsions cover the feedstock with a thin film, thus creating an immediately anaerobic environment, which further accelerates biogas production. This can also be interpreted as reduced fermenter volume requirements while maintaining the same biogas yields.

It is recommended to focus the future research not into breaking down the emulsions into O and W, but towards understanding whether the deeper disintegration could increase the reaction surface of O even more and thus further accelerate biogas production.

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