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Synthesis and characterization of release agent from used cooking oil at the concrete/formwork interface

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Abstract

For this preliminary study, the lubricating properties of six formulations containing used cooking oils were tested. The physicochemical properties involved were determined by measuring viscosities at temperatures between 0° and 40°C as well as by calculating adhesion energies. The formwork/concrete interface was then studied on the steel surface by means of both tribological measurements and analysis of facing quality at 1 day and 7 days after stripping. The results obtained were compared with a marketed vegetable-based oil. The physicochemical results revealed attractive parameters despite a viscosity that remained high. The liquid/vapor surface tensions and wettabilities measured satisfied the recommendations, i.e. greater than 30 mN/m and less than 30°. The selected formulation offered the best compromise between composition and the physicochemical recommendations. At the formwork/concrete interface and for a scraped application, satisfactory criteria were derived, with a 35% reduction in friction and an acceptable facing quality for this initial study.

Keywords : aesthetic aspect ; formulation ; friction ; lubrification ; surface tension; used cooking oil

1. Introduction

Used cooking oils (UCO) are edible oils of either vegetable origin, such as sunflower, rapeseed, and palm, or animal origin. They are used to cook food, with a change in both properties and use [1]. UCOs mainly stem from the agri-food industry, households, and the hotel, catering, and collective catering sectors [2]. It has been estimated that at least 16.54 million tons (Mt) of UCO are produced annually among the largest producing countries, namely China, Malaysia, the United States, European nations, Taiwan, Canada and Japan [1,3]. The current world production is actually estimated to lie between 20% and 32% of total vegetable oil consumption (41-67 Mt/year) [4]. In Europe, recent estimates suggest that 1.66 Mt of UCO are available, with 0.854 Mt generated from the household sector and 0.806 Mt from the commercial sector [1,4,5].

Most vegetable oils contain very high concentrations of both monounsaturated and polyunsaturated fatty acids and moreover are usually liquid at room temperature [1,3]. When these oils are heated in the presence of oxygen or of water, they undergo quite significant degradation. Oils with a high level of monounsaturated fatty acids, such as olive oil and peanut oil, are more stable and can be reused many times after heating, unlike oils with a high level of polyunsaturated fatty acids, e.g. corn oil and soybean oil [6].

In France, the collection of UCOs has become a legal obligation. As of January 1, 2012, "any person who produces or holds large quantities of waste composed mainly of biowaste, is required to set up sorting at the source and biological recovery or, when it is not carried out by a third party, a selective collection of this waste to allow for the recovery of the material in order to limit greenhouse gas emissions and encourage a return to the ground [7]. Indeed, this waste is considered non-hazardous but still cannot be mixed with household waste. In addition, used cooking oils pollute water and groundwater, clog wastewater pipes and disrupt the operation of

treatment plant operations. It is estimated that a single liter of oil can contaminate one million liters of water, thus rendering the water unfit for human and animal consumption [8].

For UCOs, restaurateurs, food industry companies, recycling centers and local authorities have since 2016 been required to submit their used cooking oil and waste to a recycling specialist once the oil volume reaches 60 liters per year. The recycling of UCO highlights a company's social and environmental responsibility while reducing both hydrocarbon consumption and greenhouse gas emissions. The combined effects of greater corporate responsibility and the legal obligation to recycle UCO now result in the vast majority of the 100,000 to 150,000 liters of used oil produced each year to be collected and recycled [9].

These oils are mainly transformed into biofuels [10,11], in particular biodiesel. It should be noted that since 2001, used oils can no longer be transformed into products intended for human or animal consumption [12]. On the other hand, cosmetics industry uses, in the form of soaps or detergents following biochemical operations [13], constitute a significant outlet for oil recovery [14,15]. In addition, a proportion of these oils is employed in producing green electricity, via adapted power plants, or else in implementing asphalt [16].

Another promising avenue for recovery would be as a release agent in the construction industry [17].

Despite recent studies [18,19] conducted on formwork surfaces without the application of formwork products, oil-based formulations are still quite widespread in both the building and prefabrication sectors. They facilitate the release of concrete, in yielding good facing quality without leaving any defects on the walls and protecting the formwork surfaces against corrosion to prolong their life. Several families of release agents can be distinguished: mineral-based, vegetable-based, and synthetic-based [20,21].

Oils derived from petrochemicals are remain the most widely used on construction sites and in prefabrication, given their technical efficiency as well as the ease of control over their formulation. However, these oils contain aromatic chemical compounds, which cause of many occupational diseases and contribute to global warming through greenhouse gas emissions. To combat these phenomena, alternatives must be sought that are both more ecological and respectful of sustainable development principles. In this aim, many applications have turned to the use of vegetable oils [22], emulsions or recycled vegetable oils. To develop these new products, it is necessary to know the physicochemical mechanisms taking place at the formwork/concrete interface. In addition to recovering UCOs via biodiesel [10,11] or industrial lubrication (in machine tools) [23], the purpose of this study is to demonstrate their potential use as a release agent. To accomplish this, it is important to analyze the physicochemical stability of UCO (viscosity, wetting, adhesion properties) and verify their ability to reduce friction in the concrete and yield high-quality facings. For this preliminary study, mixtures with additives and a solvent have been produced from UCO in the catering industry. A physicochemical study has been carried out to identify an optimal mixture. In considering the knowledge acquired by our previous research on release agents [24,25], it was advisable to introduce a streamlined application and a homogeneous film thickness to yield a dynamic viscosity below 30mPa.s, a liquid-vapor (L/V) surface tension above 30 mN/m and a drop (wetting) angle less than 30°. This research proposes an experimental approach based on our previous works [20, 21, 24-27]. Friction tests have been conducted to clarify the effectiveness of formulations derived from UCO on the concrete/oil/formwork interface. These tests make use of a plane/plane tribometer designed to study the friction of complex media. A comparison of the tribological behavior of mixtures based on both UCO and a commercial oil has also been performed.

Facing aesthetics are indeed a concern within the construction industry given that concrete walls need to reflect the quality of the completed work; these walls must not contain zero defects, such as color variation or bubbling. Facings are increasingly being left in their raw state, as the absence of finishing saves time and money. One of the parameters influencing facing quality is the release agent and especially its application. The last part of this paper will be devoted to facing quality. To better utilize the results obtained, these preliminary tests have been carried out on the steel surface. This research was conducted at the Civil Engineering Institute of Bethune (France) within the Civil Engineering and geo-Environment Laboratory (LGCgE) as part of a Master's degree internship held between February and September 2021.

2. Materials and Method

2.1. Formulations based on used cooking oil

A release agent is a mixture of various components, including the base oil, solvent, and additives (anti-corrosion, flow point depressor), aimed at achieving a high-performance formulation to ease concrete stripping without encountering any defects. For purposes of this study, the used cooking oils (UCOs) are generally composed of a mixture of edible oils, based mainly on rapeseed and sunflower. The UCO composition for this study is given in Table 1.

Table 1. Physicochemical characteristics of the used cooking oil

<i>Parameters</i>	<i>Values</i>
Acidity index (mg KOH/g)	< 1
Dynamic viscosity (Pa.s) at 20°C	0.11 ± 0.05
Average Molar Mass Triglycerides (g/mol)	> 800
Average Molar Mass Fatty Acid (g/mol)	> 200
Average length of the carbon chain C	17

119 Many solvents found in detergent formulations, cosmetics or admixtures stem from
120 petrochemicals. The goal of this work is to derive a finished product as ecological as possible
121 and the most environmentally friendly over the entire molecular scale. It is therefore preferable
122 to rely on plant esters as the solvent.

123 To complete these mixtures, various additives are needed in order to obtain a finished product
124 with the desired use properties, which may be corrosion protection, viscosity correction and anti-
125 foaming power. On the other hand, these additives can also modify the surface tension
126 characteristics and improve surface wetting properties. During the formulation step, such
127 additives can constitute up to 30% of the total mixture [28].

128 The main additives are listed below [29]:

129 - *Anti-corrosion (Ac)*

130 An anti-corrosion additive offers release agents the power to protect surfaces against corrosion
131 by forming a passivated layer; this layer is typically an oxide playing the role of a protective
132 surface film.

133 - *Pour-point depressants (PPD)*

134 Oils are known for their high sensitivity to temperature changes; as a matter of fact, at low
135 temperature they tend to gel and thicken.

136 PPD ensures that the oil remains more fluid at very low temperatures; these additives are
137 generally polymers or copolymers, such as:

138 - Copolymer olefins;

139 - Polymethacrylate polymers;

140 - Styrene-butadienes.

141

142

143

- *Thickener (T)*

Thickeners are commonly used to provide adhesion to a fluid lubricant. These additives are generally very high molecular weight polymers. The quantity to be introduced must facilitate the search for a compromise between adhesion and viscosity.

To achieve an optimal formulation that complies with the physicochemical specifications, six mixtures were prepared using used cooking oil mixed with Fatty Acid Ethyl Ester (FAEE) as the solvent and the three additives described above. Given that the proportions to be introduced were unknown for this study, it was decided to adopt average rates of anti-corrosion (Ac) and pour-point depressant (PPD), set respectively for all the mixtures at 0.1% and 0.5%. The influence of thickness is observed on the formulations Ha-1, Ha-2 and Ha-3. By taking an average value of thickener at 9%, the Hc-1, Hc-2 and Hc-3 formulations vary the solvent while trying to maintain roughly 30% additives. Table 2 details the various mixture compositions.

Table 2. Formulations of studied mixtures composed of used cooking oil

Mixtures	Ha-1	Ha-2	Ha-3	Hc-1	Hc-2	Hc-3
Oil base (H0) %	96.4	90.4	84.4	69.4	64.4	59.4
Anti-corrosion (Ac) %	0.1	0.1	0.1	0.1	0.1	0.1
Pour-point depressant (PPD) %	0.5	0.5	0.5	0.5	0.5	0.5
Solvent (FAEE) %	0	0	0	20	25	30
Thickener (T) %	3	9	15	9	9	9

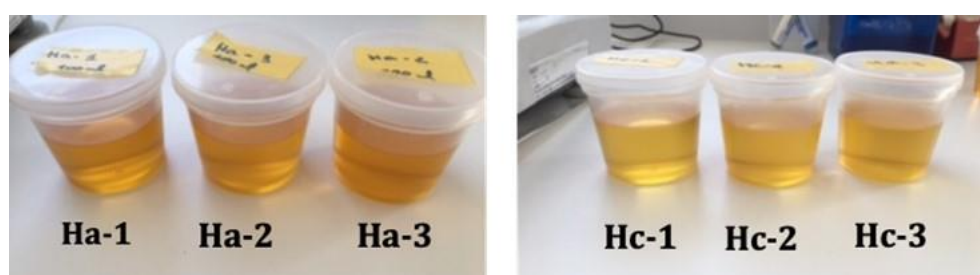


Fig. 1. Photographs of the various mixtures

The mixtures were homogenized, and Figure 1 shows the finished products. The color remains homogeneous after thermal solubilization.

To study the effect of the chemical composition of mixtures with or without a solvent, in addition to defining the role of additives and fully understanding their influence on the properties and characteristics of the ultimate mixtures, it was necessary to know their physicochemical properties, such as viscosity, wettability and liquid/vapor (L/V) surface tension. These blends were then compared to a marketed vegetable-based formulation (VO), which was pure vegetable liquid in nature, yellow to light brown in color and with a density of about 0.90.

2.2. Formulation of conventional concrete

A CEMI 52.5 CP2 type of Portland cement was used for this investigation, while the concrete employed was of a conventional variety containing 28% cement paste [25] with a mass density of 2,388 kg/m³. Workability, as measured by the Abrams cone test, is lies between 12 and 14 cm.

Table 3. Concrete mix design

Concrete	C28
Paste volume (%)	28
Cement CEMI 52,5 CP2 (kg/m ³)	232
Limestone (kg/m ³)	77
Sand 0/4 (kg/m ³)	838
Gravel 4/8 (kg/m ³)	287
Gravel 6/20 (kg/m ³)	778
Water (l)	176
W/B	0.57

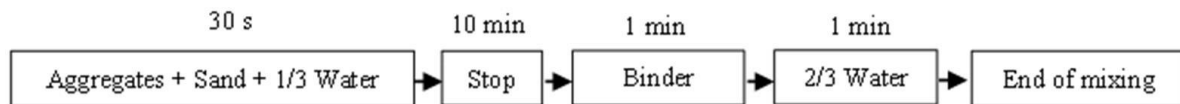
2.2.1. Implementation protocol

Metal formwork has been adopted for this study, given its extensive use within the building trades; it serves to streamline concrete setting and enhances worksite productivity. Formwork

reuse possibilities are substantial for structural elements being repeated throughout a project, e.g. vertical shear walls, floors, columns. The introduction of stiffeners enables extending elements over larger surface areas [26,27].

This new studied formwork surface was homogeneous and devoid of any defects or particularities as regards its lamination. Its roughness was measured at 1.07 μm .

Mixing was carried out in accordance with Standard NF P 18-404, entitled "Concretes: Mix design, suitability and inspection tests – Casting and storage of test specimens", which specifies the following operating procedure:



Tests were conducted in the laboratory at room temperature (around 20°C).

At construction project sites, the release agent is most often sprayed onto the formwork or spread after spraying with a rubber scraper to remove excess product specific to each individual layer. For this experimental campaign, only the application followed by scraping was studied in order to control the thickness of the film being applied.

To determine the oil film thickness before testing, pellets cut directly from a formwork were employed (Fig. 2). Spraying was performed with an Ecospray device [23] and then scraped (Fig. 2).

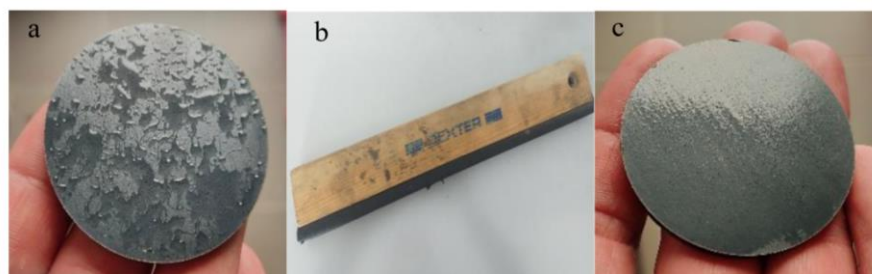


Fig 2. Sprayed Hc-2 film (a), rubber scraper (b), Scraped Hc-2 film (c),

The film thickness (e) depends on the mass (m) of product on the plate, its density (ρ), and the plate surface area (S). Weighing was performed with a precision scale of 10^{-4} g [21,26,27].

$$V = S \times e \Leftrightarrow e = \frac{V}{S}; V = \frac{m}{\rho}; \text{ then } e = \frac{m}{\rho \times S} \quad (1)$$

with S (m^2), ρ (kg/m^3), m (kg) and the thickness e (m).

Previous works have shown that values obtained by the weighing method were very close to measurements carried out by PIXE [30].

2.3. Physicochemical characterization

2.3.1. Rheological properties

The rheological properties of release agents exert a powerful influence on the behavior of the concrete/oil/formwork interface. This influence is directly felt on: wettability, friction stresses at the concrete/formwork interface, and ultimate cladding quality [24,25]. The viscosity of oils is determined using a HAAKE VT550 single-speed viscometer, fitted with a 1° (PK5) cone. A constant volume of oil ($v = 0.6 \text{ cm}^3$) is deposited on the plate with a pipette; the cone is then lowered toward the plate. To obtain an accurate measurement, a 5- to 10-min wait time (depending on measurement temperature and after depositing the oil on the plate) is required for the oil to reach the desired temperature. During the testing phase, the temperature must be held constant so as not to bias the reading. For this study, viscosity measurements were conducted at five constant temperatures (0°C , 10°C , 20°C , 30°C and 40°C).

2.3.2. Surface tension

The L/V surface tension of a liquid is correlated with the value of the contact angle formed by a drop on a support [31]. Contact angles are a readily observable physical manifestation of the more fundamental concepts of energy and surface tension. This insight serves to determine the properties of wettability and adhesion of a film, which in turn makes it possible to predict the

properties of a lining or detect the traces of surface pollutants. As part of a theoretical approach, adhesion energy values are calculated based on contact angle data.

It may be considered that the liquid/solid adhesion energy value is representative of the adhesive strength between the liquid and the support.

Young [32] was the first researcher to derive an equation for the equilibrium of a drop of liquid placed on a given solid. This resulting equation is expressed in the following form:

$$\gamma_{LV} \times \cos(\theta) = \gamma_{SV} - \gamma_{LS} \rightarrow \gamma_{LS} = \gamma_{SV} - \gamma_{LV} \times \cos(\theta) \quad (2)$$

where θ is the contact angle formed by the surface and the plane tangent to the meniscus, γ_{SV} the solid surface energy, γ_{LV} the liquid/vapor surface tension et γ_{LS} the liquid/solid interfacial tension.

The liquid/solid adhesion energy (ω_{LS}) value is also representative of the adhesive force between a liquid and its support. It is calculated according to Zisman's Equation [29]:

$$\omega_{LS} = \gamma_{LV} \times (1 + \cos(\theta)) \quad (3)$$

To determine the solid/liquid adhesion energy, it is necessary to measure both the contact angle of the oil and the liquid/vapor surface tension.

The liquid/vapor surface tension was measured using Wilhelmy's plate method at 20°C. The device employed is a tensiometer manufactured by the CAD firm fitted with a platinum blade. According to this method, a thin platinum plate is hooked up to a Kahn scale and then immersed into a liquid contained in a tank. The surface tension is indicated by the meniscus formed along the plate boundary. The measurement accuracy is ± 0.3 mN/m.

The material contact angle was determined using a PGX type of mobile goniometer. This device measures the contact angle at the interface between a liquid drop and the substrate surface where it had been deposited. The studied surface is directly obtained in a new steel formwork. The measurement accuracy is $\pm 0.3^\circ$.

2.4. Tribological study

The principle behind this type of measurement device is as follows (see Fig. 3): an interchangeable plate slides between two concrete samples, with plate movement taking place by means of a motor coupled to an auger [21,33]. The plate speed is controlled by an automaton. The samples are pressurized by activating jacks, to a pressure of between 50 and 1,000 kPa. The sample holders are cylinders fitted with a hatch for introducing the concrete. A tension sensor hooked up to the plate serves to measure the tangential friction stresses.

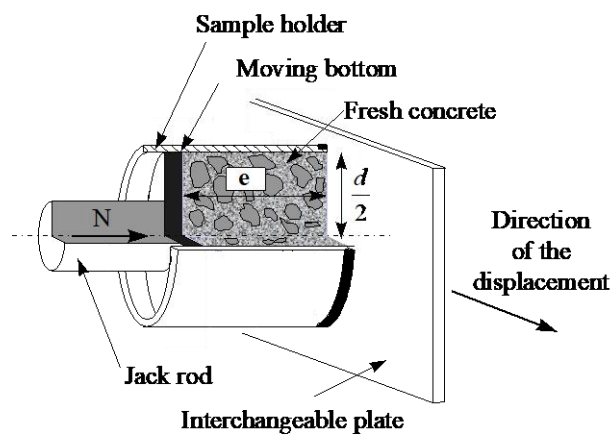


Fig. 3. Plane/Plane tribometer principle [21].

The selected parameters retained are those used in our previous studies [21,27,34] and completely mastered on the device. For our experimental campaign, these test parameters replicated jobsite conditions in order to approximate the stresses being exerted on the oil film by the concrete. The casting speed and pressures at the formwork base constitute this study's key parameters. Tests were conducted on a plate cut out from a formwork wall with roughness values of $R_a = 1.07 \mu\text{m}$. A speed of 3 m/h (0.84 mm/s) was used for our testing campaign, which corresponds to the speed commonly practiced on worksites.

Formwork heights ranging from 3 to 4.5 m have been evaluated; they correspond to pressures in the range of 75 to 115 kPa.

2.5. Aesthetic study

The cladding analysis relies on molds measuring 30*30*30 cm³ [25]. The concrete is cast in three layers and then vibrated at 5 points for 10 seconds per layer using a 40-mm diameter vibrating needle. This vibration step is carried out in five 10-second sequences per layer with the 40-mm needle.

The most significant cladding defect is a color variation, which produces a visual differentiation between large adjacent surface zones [35, 36]. This kind of alteration typically affects concrete over several millimeters of thickness. Two types of color variation exist, namely those between different concrete mixes on a single cladding, and those on the same mix. A rating scale with 7 shades of gray has been developed by CIB [37]. French Standard P18-503 also offers a guideline for evaluating the surface appearance of concrete cladding [38]. This standard is based on three characterization criteria: flatness, texture (bubbling), and hue. To analyze the cladding and formwork surfaces, the rating criteria selected were those adopted by our team when testing for the influence of mix designs on cladding quality. For this cladding, the rating was determined 24 hours after formwork removal.

The facing aspect (FA), micro-bubbling (MB), dusting (DU), dirtying (DI) and attachment points (AP) are all parameters capable of characterizing formwork removal quality [21,25]. Table 4 lists the rating criteria used.

Table 4. Grading criteria

Facing Aspect	Degree of importance	Micro-bubbling	Dusting, Dirtying, Attachment points	Degree of importance
Perfect	9	0/0.2%	None	3
Excellent	8 / 6	0.4%	Slight	2
Good	5 / 3	0.6%	A little	1
Average	2 / 0	0.8%	Normal	0
Fair	-1 / -3	1%	Many	-1
Poor	-4 / -6	1.2%	Important	-2
Very poor	-7 / -9	> 1.2%	Very important	-3

286
287 The cladding obtained was photographed on Days 1 and 7 following formwork removal. The
288 concrete blocks were released 12 hours after casting and then stored indoors at a temperature of
289 20°C +/- 2°C and constant humidity of 60% +/- 5%. The first measurement stage consisted of a
290 general assessment of the surface, complete with snapshots of its overall appearance.

291

292 3. Results and discussion

293 3.1. Viscosity analysis

294 Measurements were recorded as an average of three readings. The addition of the thickener led
295 to an increase in the dynamic viscosity of the mixtures at low temperature (Fig. 4). This variation
296 is due to the nature of vegetable oils, which are rich in vegetable esters [21]. These molecules
297 can interact with each other, hence their sensitivity to temperature changes, and such differences
298 may be restrictive when seeking to obtain a homogeneous film since the application by spraying
299 will not be completely controlled, thus making it necessary to scrape the film.

300 The presence of solvent (Fig. 5) improved the viscosity at low temperature, yielding more stable
301 values closer to those of the commercial formulation. This presence will engender a contribution
302 beneficial to the application. The viscosity value remains high compared to the recommendation
303 of 30 mPa.s at 20°C [25]. The viscosity values found in the literature lie between 0.02 Pa.s and
304 0.05 Pa.s [25, 39-41]. However, Hashin Mohseni's [42] and Loh's [43] research on vegetable oils
305 (palm) demonstrated an improved mold release functionality due to its higher viscosity qualities.
306 This analysis is encouraging for the pursuit of this study.

307 Continuing the analysis makes it possible to derive the interfacial parameters necessary for an
308 improved adhesion of the oil on the formwork surface.

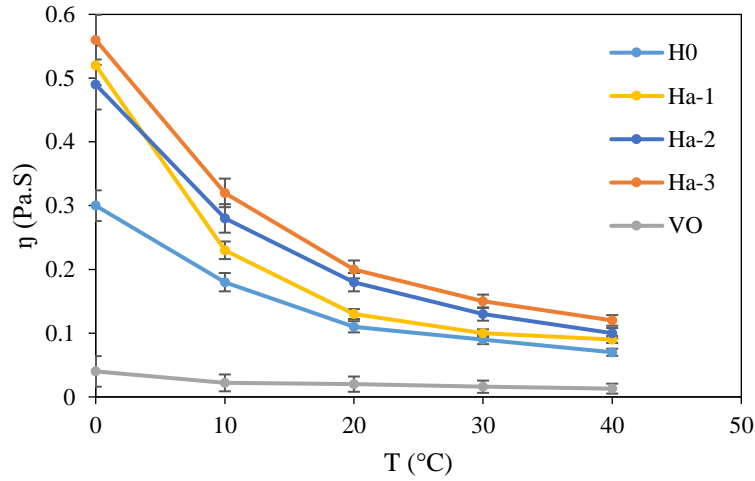


Fig. 4. Dynamic viscosities of mixtures without solvent as a function of temperature

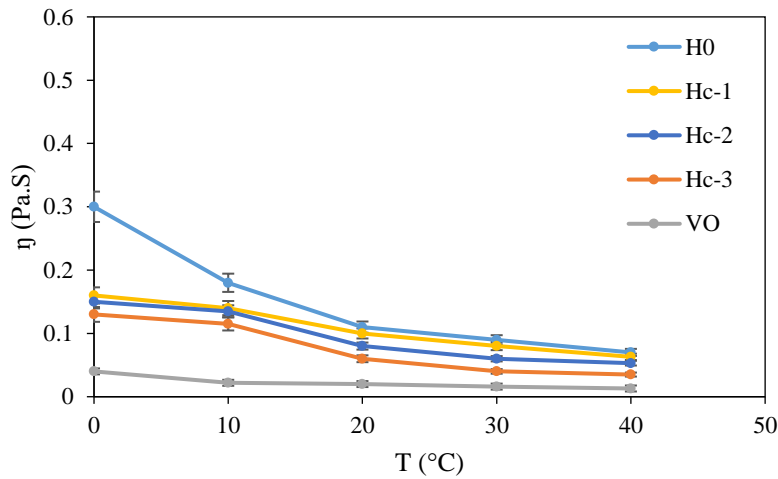


Fig. 5. Dynamic viscosities of mixtures with solvent as a function of temperature

3.2. Surface tension analysis

The corresponding values are given in Table 4. The liquid/vapor surface tensions lie between 34 and 37 mN/m, which correspond to the recommendations promulgated for other release agents [22]. It had previously been shown that a minimum value of 30 mN/m made it possible to obtain effective adhesion energy.

Courard *et al.* [44] reached the same conclusions regarding interfacial properties, i.e. low wettability and high surface tension on steel and plywood formwork surfaces. Results indicate

that adding the thickener is favorable to the L/V surface tension yet degrades wettability. The presence of solvent acts upon the wettability of mixtures, making it possible to retain a good amount of adhesion energy. The mixtures obtained display a higher adhesion energy value than that of the commercial vegetable formulation. In a previous study [45] on recycled vegetable-based oils, the presence of solvent had influenced the viscosity in yielding values close to those of a commercial formulation while maintaining stable surface tension properties. The solvent did not improve facing quality.

In considering physicochemical results, the study continues with the Hc-2 formulation, which allows for a compromise between a large quantity of base cooking oil used and the solvent, thereby both stabilizing viscosity and reducing wettability.

Table 4. Interface parameter values

	$\gamma_{L/V}$ (mN/m)	θ (°)	$\omega_{L/S}$ (mN/m)
H0	34.5	18.8	67.2
Ha-1	34.9	23.1	67.0
Ha-2	35.7	24.9	68.1
Ha-3	36.9	25.6	70.2
Hc-1	35.5	18.3	69.2
Hc-2	35.1	18.1	68.5
Hc-3	34.6	17.4	67.6
VO	33.5	15.2	65.8

3.3. Influence of UCOs at the formwork/concrete interface

The weighing measurements provided the oil film thickness values, which lie between 5 and 7 μm for the Hc-2 formulation and between 3 and 5 μm for the vegetable oil (VO).

3.3.1. Friction analysis

To reproduce the implementation of the concrete in the formwork, tests were carried out with a plane/plane tribometer [21,28,32]. Such tests served to validate the mechanisms at the

formwork/concrete interface due to the presence of a physical effect or a chemical effect [17]. The Hc-2 formulation was to be compared with the marketed vegetable-based formulation (VO). The evolution in the friction constraints of the concrete and the demolding oil films is displayed in Figure 6.

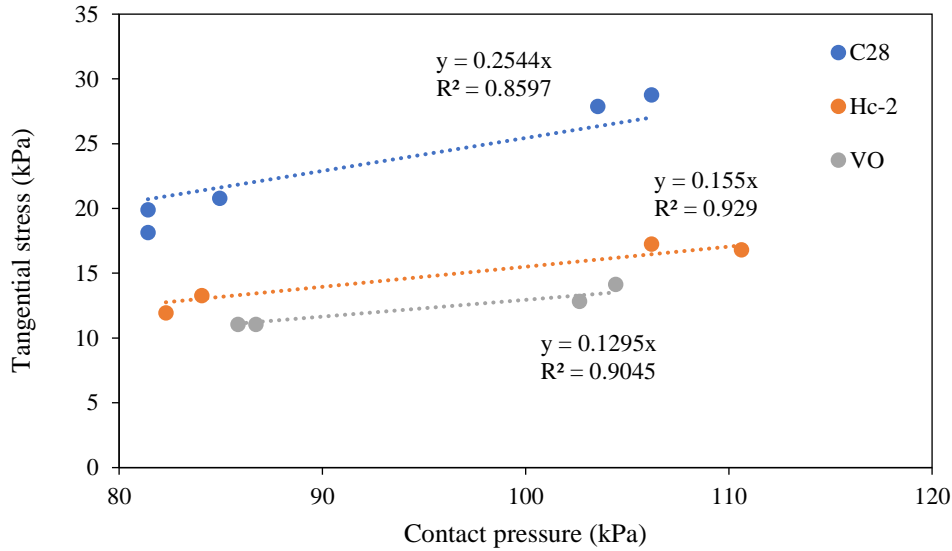


Fig. 6. Friction stresses as a function of contact pressure

For this measurement range, the concrete is submitted to the following Coulomb Law:

$$\tau_f = \mu \times P \quad (4)$$

with τ_f the tangential stress (Pa) and P the contact pressure (Pa).

The results in Figure 6 show great efficiency of the Hc-2 mixture in reducing concrete friction: the Hc-2 mixture reduces concrete friction by an average of 35% vs. roughly 45% for the vegetable formulation (VO). These results are similar to those reported in previous studies [21,28,32]. Leon-Martinez *et al.* [40] showed similar results on refined oils. Their measurements revealed a reduction in tangential adhesion of 64% compared to a surface free of a release agent. Bouharoun *et al.* [39] also exposed the effectiveness of vegetable oils in reducing the friction of self-compacting concrete. For comparison in the field of industrial lubrication [46-48], formulations based on vegetable oil are often studied and exhibit good lubrication performance. All these studies were carried out on unprocessed products, hence the great interest in pursuing

this study on UCOs. In addition, Reeves *et al.* [49] demonstrated that lubrication performance is correlated with viscosity and depends on the nature of the oils. Commercial oil remains more effective with a lower applied thickness. However, the Hc-2 formulation has a thicker film and appears to be more cohesive on the surface. Its high adhesion energy can also pose a constraint on the lubricating effect of the film and therefore limit its effectiveness on friction. These analyses have allowed us to add a criterion to our specifications, namely an adhesion energy of less than 65 mN/m, along with an L/V surface tension above 30 mN/m and a drop angle smaller than 30°.

To complete the action process at the formwork/concrete interface, the slopes of the two straight lines seem to be similar and not parallel to the concrete, which would indicate that UCOs form a double layer (esters and soap), as is typical of vegetable-based formulations. The physical and chemical effects are therefore combined (Fig. 7), as has been demonstrated in previous studies [21] and confirmed by the research of both Leon-Martinez *et al.* [40] and Loh *et al.* [43]. The film is thus more resistant and effective during the passage of concrete.

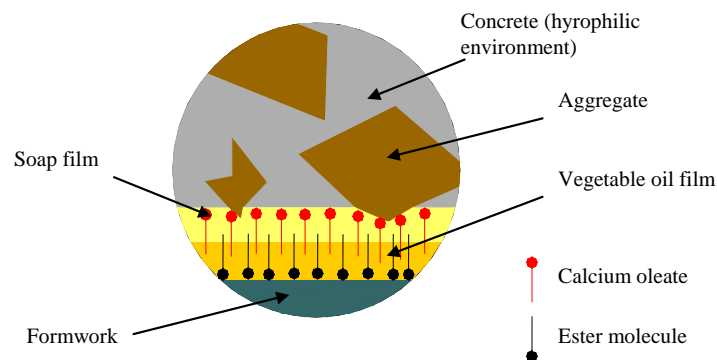


Fig. 7. Organization of the concrete/formwork interface in the presence of vegetable oil [21]

To finalize this initial approach, the facing aesthetics will now be analyzed.

3.3.2. Analysis of surfaces of molds and concrete facings

Figure 8 shows the surface condition of the mold after stripping. The surface of the formwork with Hc-2 contains more dirt and sticking points; it remains "fatty" and is difficult to clean compared to the commercial formulation (VO).



Fig. 8. Surface condition of the molds after stripping: VO on the left, Hc-2 on the right

Figures 9 and 10 display the concrete facings obtained 1 day and 7 days after stripping. The facing evolves positively over time, in recognition of the utility often demonstrated when waiting 7 days to validate the quality of a concrete facing [21,37]. Similar and interesting results were obtained from feasibility studies to develop palm olein-based lubricants with a tackifier [43] or edible oil-based release oils [41] with respect to both dismantling and the performance on finished concrete blocks. These outcomes were of higher quality than the facings obtained during a previous study on recycled vegetable industrial oils [45].



Fig. 9. Concrete facing obtained after 1 day: VO on the left, Hc-2 on the right



Fig. 10. Concrete facing obtained after 7 days: VO on the left, Hc-2 on the right

Table 5 summarizes results according to the observed criteria. The notation used is the average of 4 surfaces, as derived by two observers. The concrete surfaces reveal little micro-bubbling and dusting. Micro-bubbling was determined by means of image analysis [45]. Facing quality improved at 7 days. The phenomenon of concrete maturity [50] leads to an evolution in its color; in fact, the concrete tends to lighten over time. The Hc-2 mixture mainly loses points on formwork surface quality due to a greater presence of fouling.

Table 5. Notation of formwork surfaces and concrete facings

	Concrete facing						Mold surfaces				Total	Total
	FA	FA	MB	DU	Total /15	Total /15	DI	DU	AP	Total	/24	Total
	(1day)	(7days)			(1day)	(7days)				/9	(1day)	(7days)
Steel surface (spray application + scraping)												
VO	6	8	2	2	10	12	2	2	1	5	15	17
Hc-2	4	7	2	1	7	10	1	1	1	3	10	13

4. Conclusion

This initial study of mixtures based on used cooking oil (UCO) as a mold release agent has yielded noteworthy conclusions as well as prospects for further research. First, the physicochemical characterizations showed that the formulated products can behave similarly to commercial formulations. However, the products still need to be optimized in order to improve

viscosity and facilitate application. The addition of a thickener is not currently recommended for UCOs.

Indeed, application by spraying alone has not yet been mastered at this stage of the study. The interfacial parameters potentially stand out, given that the tribometer measurements indicate a higher frictional resistance for the Hc-2 oil film (6 μm thick). Film thickness therefore is not the main parameter associated with this study, and moreover it is recommended to limit the adhesion energy to a maximum of 65 mN/m. In addition, vegetable-based formulations involve more of a chemical process by creating a saponification film at the interface. Consequently, it is critical to control the quantity deposited in order to avoid excessive dusting or dirtying.

An analysis in the presence of concrete has offered positive conclusions regarding the friction reduction of the selected Hc-2 product for a scraped application after spraying. The chemical effect is predominant and allowed for effective lubrication. The concrete facings obtained were of good quality at 7 days; however, as mentioned above, the mold surface remained heavily clogged and difficult to clean.

Following this study, it will be necessary to refine the Hc-2 mixture from a physicochemical standpoint to both improve its application (through spraying alone) and verify its stability over time. It would also be worthwhile to quantify the performance of the product relative to steel formwork corrosion by means of electrochemical impedance spectroscopy measurements. It would indeed be interesting to compare the results by measurements on wooden surfaces. Lastly, tests on building sites would need to be carried out to validate its marketing feasibility.

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Conflict of interest

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References

- [1] Loizides MI, Loizidou XI, Orthodoxou DL, Petsa D (2019) Circular Bioeconomy in Action: Collection and Recycling of Domestic Used Cooking Oil through a Social, Reverse Logistics System. Recycling. 4:16. <https://doi.org/10.3390/recycling4020016>
- [2] Orjuela A, Clark J (2020) Green chemicals from used cooking oils: Trends, challenges, and opportunities. Green Sustain Chem 26:100369. <https://doi.org/10.1016/j.cogsc.2020.100369>
- [3] Gui MM, Lee KT, Bhatia S (2008) Feasibility of edible oil vs. non-edible oil vs. waste edible oil as biodiesel feedstock. Energy 33:1646–1653. <https://doi.org/10.1016/j.energy.2008.06.002>
- [4] Ribau M, Nogueira R, Miguel L (2018) Quantitative assessment of the valorization of used cooking oils in 23 countries. Waste Manag. 78:611–620. <https://doi.org/10.1016/j.wasman.2018.06.039>
- [5] Greenea (2018) Analysis of the current development of household UCO collection systems in the EU.

462 <https://www.theicct.org/sites/default/files/publications/Greenea%20Report%20Household%20>
463 [UCO%20Collection%20in%20the%20EU_ICCT_20160629.pdf](https://www.theicct.org/sites/default/files/publications/Greenea%20Report%20Household%20UCO%20Collection%20in%20the%20EU_ICCT_20160629.pdf). Accessed 15 November 2018
464 [6] Ammar C, Amor S (2022) Stability and Change in Fatty Acids Composition of Soybean,
465 Corn, and Sunflower Oils during the Heating Process. J Food Qual 17p.
466 <https://doi.org/10.1155/2022/6761029>
467 [7] Walker DB, Baumgartner DJ, Gerba CP, Fitzsimmons K (2019) Chapter 16 - Surface Water
468 Pollution. In: Mark L. Brusseau, Ian L. Pepper, Charles P. Gerba (ed) Environmental and
469 Pollution Science, 3rd edn. Academic Press, pp261-292
470 [8] Environmental Code - Circular of 10/01/12 relating to the methods of application of the
471 obligation to sort bio-waste at source by large producers - article L 541-21-1, France
472 [9] Choe E, Min DB, (2007) Chemistry of deep-fat frying oils. J Food Sci 72:77–86.
473 <https://doi.org/10.1111/j.1750-3841.2007.00352.x>
474 [10] Fonseca JM, Teleken JG, de Cinque Almeida V, da Silva C (2019) Biodiesel from waste
475 frying oils: Methods of production and purification. Energy Convers Manag 184:205-218.
476 <https://doi.org/10.1016/j.enconman.2019.01.061>
477 [11] Kumar S, Sushma U, Chandrasagar L, Raju V, Devi V (2017) Use of Waste Frying Oil as
478 C.I. Engine Fuel—A Review. Open Access Library Journal 4:1-33.
479 <https://doi.org/10.4236/oalib.1103958>
480 [12] Gui MM, Lee KT, Bhatia S (2008) Feasibility of edible oil vs. non-edible oil vs. waste edible
481 oil as biodiesel feedstock. Energy 33:1646–1653. <https://doi.org/10.1016/j.energy.2008.06.002>
482 [13] Sajid M, Ahmad Khan MS, Singh Cameotra S, Safar Al-Thubiani A (2020) Biosurfactants:
483 Potential applications as immuno-modulator drugs. Immunol Lett 223:71-77.
484 <https://doi.org/10.1016/j.imlet.2020.04.003>

485 [14] Frota de Albuquerque Landi F, Fabiani C, Castellani B, Cotana F, Pisello AL (2022)
 486 Environmental assessment of four waste cooking oil valorization pathways. Waste Manage
 487 138:219-233. <https://doi.org/10.1016/j.wasman.2021.11.037>

488 [15] Kim JH, Oh YR, Hwang J et al (2021) Valorization of waste-cooking oil into sophorolipids
 489 and application of their methyl hydroxyl branched fatty acid derivatives to produce engineering
 490 bioplastics. Waste Manage 124:195-202. <https://doi.org/10.1016/j.wasman.2021.02.003>

491 [16] Aghazadeh Dokandari P, Kaya D, Sengoz B, Topal A Implementing waste oils with
 492 reclaimed asphalt pavement, Proceedings of the 2nd World Congress on Civil, Structural, and
 493 Environmental Engineering (CSEE'17) Barcelona, Spain, April 2 – 4, 2017

494 [17] de Brito J, dos Santos R, Branco FA, (2000) Evaluation of the technical performance of
 495 concrete vegetable oil-based release agents. Mater Struct 33:262–269

496 [18] Spitz N, Coniglio N, El Mansori M, Montagne A, Mezghani S (2018) On functional
 497 signatures of bare and coated formwork skin surfaces. Constr Build Mater 189:560–567.
 498 <https://doi.org/10.1016/j.conbuildmat.2018.09.042>

499 [19] Chadfeau C, Omary S, Belhaj E, Fond C, Feugeas F (2021) Characterization of the surface
 500 of formworks – Influence of the surface energy and surface texture parameters on the demolding
 501 forces. Constr Build Mater 272 :121947. <https://doi.org/10.1016/j.conbuildmat.2020.121947>

502 [20] de Caro P, Djelal C, Libessart L, Dubois I (2007) Influence of the nature of demoulding
 503 agent on the properties of the formwork/concrete interface. Mag Concr Res 59:141-149.
 504 <https://doi.org/10.1680/mac.2007.59.2.141>

505 [21] Djelal C, de Caro P, Libessart L, Dubois I, Pébère N (2008) Comprehension of demoulding
 506 mechanics at the formwork/oil/concrete interface. Mater Struct 41:571-581.
 507 <https://doi.org/10.1617/s11527-007-9268-3>

508 [22] Uppar R, Dinesha P, Kuma S (2022) A critical review on vegetable oil-based bio-lubricants:
509 preparation, characterization, and challenges. Environ Dev
510 Sustain <https://doi.org/10.1007/s10668-022-02669-w>

511 [23] Mannu A, Garroni S, Ibanez Porras J, Mele A (2020) Available Technologies and Materials
512 for Waste Cooking Oil Recycling. Processes 8(3):366. <https://doi.org/10.3390/pr8030366>

513 [24] Libessart L, de Caro P, Djelal C, Dubois I (2015) Correlation between adhesion energy of
514 release agents on the formwork and demoulding performances. Constr Build Mater 76:130-139
515 <https://doi.org/10.1016/j.conbuildmat.2014.11.061>

516 [25] Libessart L, Djelal C, de Caro P (2014) Influence of the type of release oil on steel formwork
517 corrosion and facing aesthetics. Constr Build Mater 68:391-401.
518 <https://doi.org/10.1016/j.conbuildmat.2014.06.065>

519 [26] Libessart L, Lahlou A, Djelal C, Vanhove Y, Mateo S (2018) Influence of the emulsions to
520 the interface formwork/concrete. ACI - Special Publication 326:1-10.
521 <https://doi.org/10.14359/51711106>

522 [27] Libessart L, Djelal C, de Caro P, Laiymani I (2020) Comparative study of the tribological
523 behaviour of emulsions and demoulding oils at the concrete/formwork interface. Constr Build
524 Mater 239:117826. <https://doi.org/10.1016/j.conbuildmat.2019.117826>

525 [28] Machinery Lubrication (2018) Lubricant Additives - A Practical Guide
526 <https://www.machinerylubrication.com/Read/31107/oil-lubricant-additives> , Noria Corporation

527 [29] Mechanic (2018) A Practical Guide To Understanding Engine Oil Additives.
528 <https://trade.mechanic.com.au/news/a-practical-guide-to-understanding-engine-oil-additives>.
529 Accessed 28 August 2018

530 [30] Djelal C, Vanhove Y, Chambellan D, Brisset P (2010) Influence of the thickness of
531 demoulding oils on the aesthetic quality of facings. Mater Struct 43:687-698

532 [31] Chappuis J, Georges JM (1974) Contribution to Study of Wetting - Analysis of a Measuring
533 Method. *Phys Phys Chim Biol.* 71:567-575

534 [32] Zisman WA (1964) Relation of the Equilibrium Contact angle to Liquid and Solid
535 Constitution. *Adv Chem* 43:1-51

536 [33] Djelal C, Vanhove Y, Libessart L (2016) Analysis of friction and lubrication conditions of
537 concrete/formwork interfaces. *Int J Civ Eng Tech* 7:18-30

538 [34] Spitz N, Coniglio N, Libessart L, El Mansori M, Djelal C (2021) Characterizing tribological
539 behavior of fresh concrete against formwork surfaces. *Constr Build Mater* 303 :124233.
540 <https://doi.org/10.1016/j.conbuildmat.2021.124233> ,

541 [35] Collection technique CimBéton (2005) Bétons et ouvrages d’art – La maîtrise esthétique des
542 parements en béton. T49. France

543 [36] Lemaire G, Escadeillas G, Ringot E (2005) Evaluating concrete surfaces using an image
544 analysis process. *Constr Build Mater* 19:604–611.
545 <https://doi.org/10.1016/j.conbuildmat.2005.01.025>

546 [37] CIB (1973) Tolérances sur les défauts d’aspect du béton. Rapport n° 24 – n°1189

547 [38] ST P18-503, “Concrete surfaces and facing. Identification elements”, ISSN:0335-3931,
548 1989

549 [39] Bouharoun S, de Caro P, Dubois I, Djelal C, Vanhove Y (2013) Effect of a superplasticizer
550 on the properties of the concrete/oil/formwork interface. *Constr Build Mater* 47:1137-1144
551 <https://doi.org/10.1016/j.conbuildmat.2013.05.029>

552 [40] León-Martínez FM, Abad-Zarate EF, Lagunez-Rivera L et al (2016) Laboratory and field
553 performance of biodegradable release agents for hydraulic concrete. *Mater Struct* 49: 2731–2748.
554 <https://doi.org/10.1617/s11527-015-0681-8>

555 [41] Barnat-Hunek D, Szafraniec M (2021) Influence of biodegradable release oils on the
 556 physical and mechanical properties of light-colored architectural concrete. *Materials* 14:4630.
 557 <https://doi.org/10.3390/ma14164630>

558 [42] Mohseni SH, Bahij S, Omary S, Feugeas F, Birinci F (2022) Effect of formwork surface
 559 texture features on surface morphology, roughness parameters, and the demolding force of
 560 cementitious materials. *J Civil Environ Eng* 12(5)

561 [43] Loh SH, Muzzammil N (2022) Palm olein lubricates and removes freshly baked concrete
 562 from its formwork. *Journal of Oil Palm Research* 34:323-336.
 563 <https://doi.org/10.21894/jopr.2021.0038>

564 [44] Courard L, Goffinet C, Migeotte N et al (2012) Influence of the reuse of OSB and marine
 565 plywood formworks on concrete surface aesthetics. *Mater Struct* 45:331–1343.
 566 <https://doi.org/10.1617/s11527-012-9835-0>

567 [45] Woma TY, Lawal SA, Abdulrahman AS, Olutoye MA, Ojapah MM (2019) Vegetable Oil
 568 Based Lubricants: Challenges and Prospects. *Tribology Online* 14(2):60-70
 569 <https://doi.org/10.2474/trol.14.60>

570 [46] Bahari A, Lewis R, Slatter T (2018) Friction and wear response of vegetable oils and their
 571 blends with mineral engine oil in a reciprocating sliding contact at severe contact conditions. *P I*
 572 *MECH ENG J-J ENG* 232(3):244-258 <https://doi.org/10.1177/1350650117712344>

573 [47] Pawar RV, Hulwan DB, Mandale MB (2022) Recent advancements in synthesis, rheological
 574 characterization, and tribological performance of vegetable oil-based lubricants enhanced with
 575 nanoparticles for sustainable lubrication, *J Clean Prod* 378:134454
 576 <https://doi.org/10.1016/j.jclepro.2022.134454>

577 [48] Reeves CJ, Menezes PL, Jen T-C, Lovell MR (2015) The influence of fatty acids on
 578 tribological and thermal properties of natural oils as sustainable biolubricants, *Tribol Int* 90:123-
 579 134 <https://doi.org/10.1016/j.triboint.2015.04.021>

580 [50] Lukpanov R, Zharassov S, Mkilima T, Aldungarova A (2021) Performance of maturity
581 method for estimation of concrete strength based on cubic specimens. Technobius 1:4 0008.
582 <https://doi.org/10.54355/tbus/1.4.2021.0008>