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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 ; lubrication ; surface tension; used cooking oil

27 1. Introduction

28 Used cooking oils (UCO) are edible oils of either vegetable origin, such as sunflower, rapeseed,
29 and palm, or animal origin. They are used to cook food, with a change in both properties and use
30 [1]. UCOs mainly stem from the agri-food industry, households, and the hotel, catering, and
31 collective catering sectors [2]. It has been estimated that at least 16.54 million tons (Mt) of UCO
32 are produced annually among the largest producing countries, namely China, Malaysia, the
33 United States, European nations, Taiwan, Canada and Japan [1,3]. The current world production
34 is actually estimated to lie between 20% and 32% of total vegetable oil consumption (41-67
35 Mt/year) [4]. In Europe, recent estimates suggest that 1.66 Mt of UCO are available, with 0.854
36 Mt generated from the household sector and 0.806 Mt from the commercial sector [1,4,5].

37
38 Most vegetable oils contain very high concentrations of both monounsaturated and
39 polyunsaturated fatty acids and moreover are usually liquid at room temperature [1,3]. When
40 these oils are heated in the presence of oxygen or of water, they undergo quite significant
41 degradation. Oils with a high level of monounsaturated fatty acids, such as olive oil and peanut
42 oil, are more stable and can be reused many times after heating, unlike oils with a high level of
43 polyunsaturated fatty acids, e.g. corn oil and soybean oil [6].

44
45 In France, the collection of UCOs has become a legal obligation. As of January 1, 2012, "any
46 person who produces or holds large quantities of waste composed mainly of biowaste, is required
47 to set up sorting at the source and biological recovery or, when it is not carried out by a third
48 party, a selective collection of this waste to allow for the recovery of the material in order to limit
49 greenhouse gas emissions and encourage a return to the ground [7]. Indeed, this waste is
50 considered non-hazardous but still cannot be mixed with household waste. In addition, used
51 cooking oils pollute water and groundwater, clog wastewater pipes and disrupt the operation of

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

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

61
62 These oils are mainly transformed into biofuels [10,11], in particular biodiesel. It should be noted
63 that since 2001, used oils can no longer be transformed into products intended for human or
64 animal consumption [12]. On the other hand, cosmetics industry uses, in the form of soaps or
65 detergents following biochemical operations [13], constitute a significant outlet for oil recovery
66 [14,15]. In addition, a proportion of these oils is employed in producing green electricity, via
67 adapted power plants, or else in implementing asphalt [16].

68 Another promising avenue for recovery would be as a release agent in the construction industry
69 [17].

70
71 Despite recent studies [18,19] conducted on formwork surfaces without the application of
72 formwork products, oil-based formulations are still quite widespread in both the building and
73 prefabrication sectors. They facilitate the release of concrete, in yielding good facing quality
74 without leaving any defects on the walls and protecting the formwork surfaces against corrosion
75 to prolong their life. Several families of release agents can be distinguished: mineral-based,
76 vegetable-based, and synthetic-based [20,21].

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

100

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

110 2. Materials and Method

111 2.1. Formulations based on used cooking oil

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

117 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

118

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

144 - *Thickener (T)*

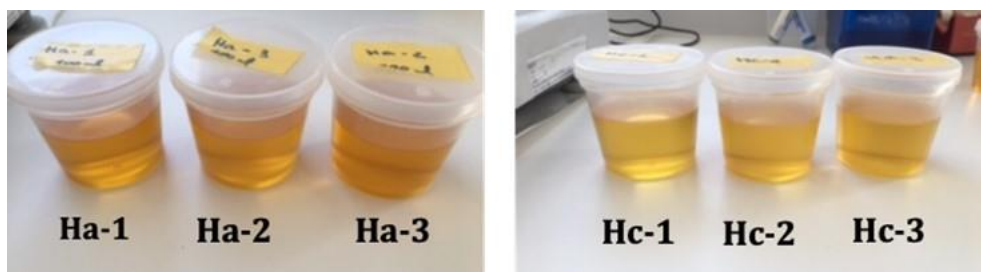
145 Thickeners are commonly used to provide adhesion to a fluid lubricant. These additives are
146 generally very high molecular weight polymers. The quantity to be introduced must facilitate the
147 search for a compromise between adhesion and viscosity.

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

156 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

157



158

159

Fig. 1. Photographs of the various mixtures

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

162
163 To study the effect of the chemical composition of mixtures with or without a solvent, in addition
164 to defining the role of additives and fully understanding their influence on the properties and
165 characteristics of the ultimate mixtures, it was necessary to know their physicochemical
166 properties, such as viscosity, wettability and liquid/vapor (L/V) surface tension. These blends
167 were then compared to a marketed vegetable-based formulation (VO), which was pure vegetable
168 liquid in nature, yellow to light brown in color and with a density of about 0.90.

169

170 2.2. Formulation of conventional concrete

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

174 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

175

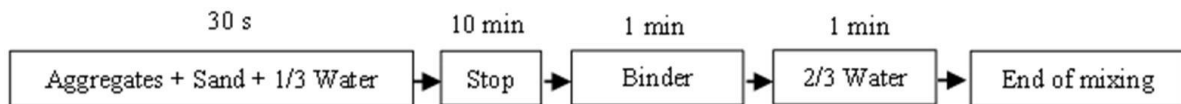
176 2.2.1. Implementation protocol

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

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

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

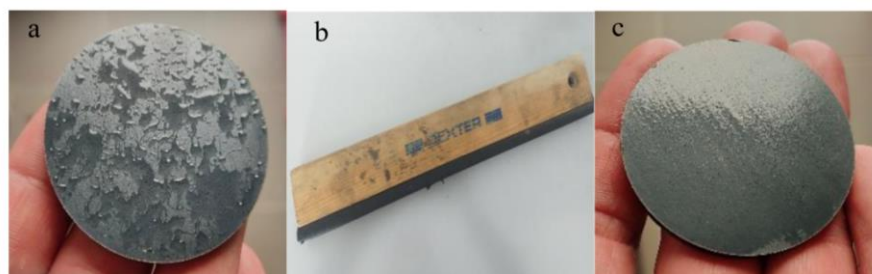
184 Mixing was carried out in accordance with Standard NF P 18-404, entitled "Concretes: Mix
185 design, suitability and inspection tests – Casting and storage of test specimens", which specifies
186 the following operating procedure:



187
188 Tests were conducted in the laboratory at room temperature (around 20°C).

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

193 To determine the oil film thickness before testing, pellets cut directly from a formwork were
194 employed (Fig. 2). Spraying was performed with an Ecospray device [23] and then scraped (Fig.
195 2).



196
197 Fig 2. Sprayed Hc-2 film (a), rubber scraper (b), Scraped Hc-2 film (c),

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

$$200 \quad V = S \times e \Leftrightarrow e = \frac{V}{S}; \quad V = \frac{m}{\rho}; \quad \text{then} \quad e = \frac{m}{\rho \times S} \quad (1)$$

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

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

204

205 2.3. Physicochemical characterization

206 2.3.1. Rheological properties

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

217 2.3.2. Surface tension

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

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

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

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

$$228 \quad \gamma_{LV} \times \cos(\theta) = \gamma_{SV} - \gamma_{LS} \rightarrow \gamma_{LS} = \gamma_{SV} - \gamma_{LV} \times \cos(\theta) \quad (2)$$

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

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

$$233 \quad \omega_{LS} = \gamma_{LV} \times (1 + \cos(\theta)) \quad (3)$$

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

236 The liquid/vapor surface tension was measured using Wilhelmy's plate method at 20°C. The
237 device employed is a tensiometer manufactured by the CAD firm fitted with a platinum blade.

238 According to this method, a thin platinum plate is hooked up to a Kahn scale and then immersed
239 into a liquid contained in a tank. The surface tension is indicated by the meniscus formed along
240 the plate boundary. The measurement accuracy is ± 0.3 mN/m.

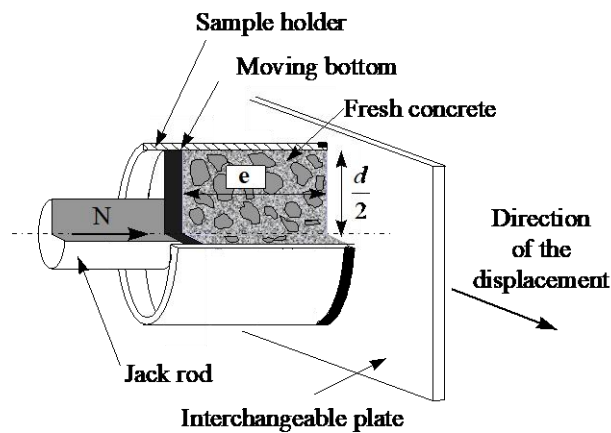
241 The material contact angle was determined using a PGX type of mobile goniometer. This device
242 measures the contact angle at the interface between a liquid drop and the substrate surface where
243 it had been deposited. The studied surface is directly obtained in a new steel formwork. The
244 measurement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

245

246

247 2.4. Tribological study

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



254
255 Fig. 3. Plane/Plane tribometer principle [21].

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

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

265
266

267 2.5. Aesthetic study

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

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

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

285 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 $20^{\circ}\text{C} \pm 2^{\circ}\text{C}$ and constant humidity of $60\% \pm 5\%$. The first measurement stage consisted of a
290 general assessment of the surface, complete with snapshots of its overall appearance.

291

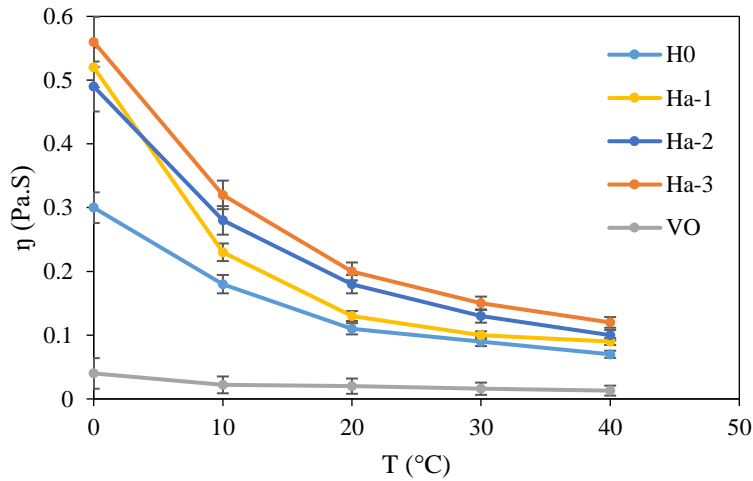
292 3. Results and discussion

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.

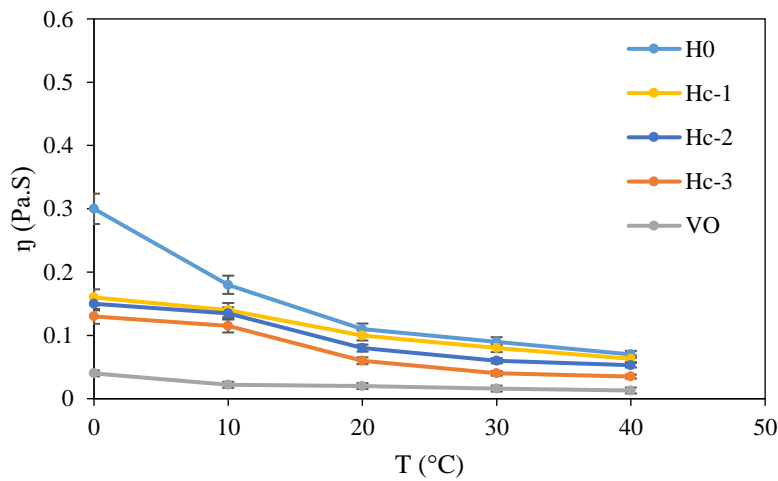
300 The presence of solvent (Fig. 5) improved the viscosity at low temperature, yielding more stable
301 values closer to those of the commercial formulation. This presence will engender a contribution
302 beneficial to the application. The viscosity value remains high compared to the recommendation
303 of $30 \text{ mPa}\cdot\text{s}$ at 20°C [25]. The viscosity values found in the literature lie between $0.02 \text{ Pa}\cdot\text{s}$ and
304 $0.05 \text{ Pa}\cdot\text{s}$ [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.



309

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



311

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

313

314 3.2. Surface tension analysis

315 The corresponding values are given in Table 4. The liquid/vapor surface tensions lie between 34
 316 and 37 mN/m, which correspond to the recommendations promulgated for other release agents
 317 [22]. It had previously been shown that a minimum value of 30 mN/m made it possible to obtain
 318 effective adhesion energy.

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

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

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

331 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

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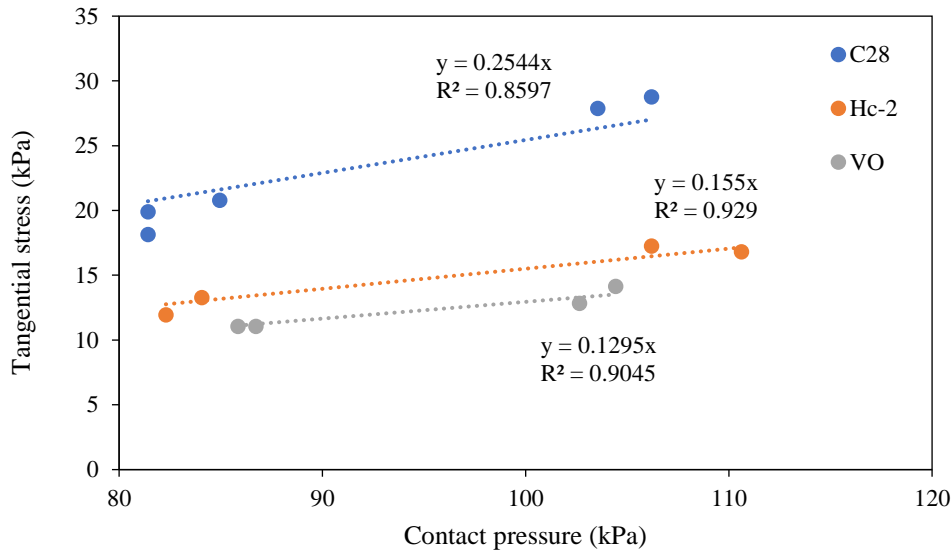
333 3.3. Influence of UCOs at the formwork/concrete interface

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

336 3.3.1. Friction analysis

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

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



343

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

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

346
$$\tau_f = \mu \times P \tag{4}$$

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

348 The results in Figure 6 show great efficiency of the Hc-2 mixture in reducing concrete friction:

349 the Hc-2 mixture reduces concrete friction by an average of 35% vs. roughly 45% for the

350 vegetable formulation (VO). These results are similar to those reported in previous studies

351 [21,28,32]. Leon-Martinez *et al.* [40] showed similar results on refined oils. Their measurements

352 revealed a reduction in tangential adhesion of 64% compared to a surface free of a release agent.

353 Bouharoun *et al.* [39] also exposed the effectiveness of vegetable oils in reducing the friction of

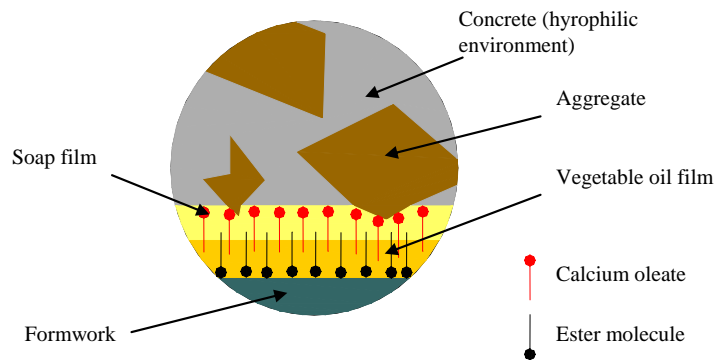
354 self-compacting concrete. For comparison in the field of industrial lubrication [46-48],

355 formulations based on vegetable oil are often studied and exhibit good lubrication performance.

356 All these studies were carried out on unprocessed products, hence the great interest in pursuing

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

365 To complete the action process at the formwork/concrete interface, the slopes of the two straight
366 lines seem to be similar and not parallel to the concrete, which would indicate that UCOs form a
367 double layer (esters and soap), as is typical of vegetable-based formulations. The physical and
368 chemical effects are therefore combined (Fig. 7), as has been demonstrated in previous studies
369 [21] and confirmed by the research of both Leon-Martinez *et al.* [40] and Loh *et al.* [43]. The
370 film is thus more resistant and effective during the passage of concrete.



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

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

379
380
381

382 3.3.2. Analysis of surfaces of molds and concrete facings

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



386

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

388 Figures 9 and 10 display the concrete facings obtained 1 day and 7 days after stripping. The
389 facing evolves positively over time, in recognition of the utility often demonstrated when waiting
390 7 days to validate the quality of a concrete facing [21,37]. Similar and interesting results were
391 obtained from feasibility studies to develop palm olein-based lubricants with a tackifier [43] or
392 edible oil-based release oils [41] with respect to both dismantling and the performance on
393 finished concrete blocks. These outcomes were of higher quality than the facings obtained during
394 a previous study on recycled vegetable industrial oils [45].

395



396

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



397

398

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

399

Table 5 summarizes results according to the observed criteria. The notation used is the average

400

of 4 surfaces, as derived by two observers. The concrete surfaces reveal little micro-bubbling and

401

dusting. Micro-bubbling was determined by means of image analysis [45]. Facing quality

402

improved at 7 days. The phenomenon of concrete maturity [50] leads to an evolution in its color;

403

in fact, the concrete tends to lighten over time. The Hc-2 mixture mainly loses points on

404

formwork surface quality due to a greater presence of fouling.

405

Table 5. Notation of formwork surfaces and concrete facings

	Concrete facing						Mold surfaces				Total	Total
	FA (1day)	FA (7days)	MB	DU	Total /15 (1day)	Total /15 (7days)	DI	DU	AP	Total /9	/24 (1day)	/24 (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

406

407 4. Conclusion

408

This initial study of mixtures based on used cooking oil (UCO) as a mold release agent has

409

yielded noteworthy conclusions as well as prospects for further research. First, the

410

physicochemical characterizations showed that the formulated products can behave similarly to

411

commercial formulations. However, the products still need to be optimized in order to improve

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

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

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

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

432

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435

436

437 Conflict of interest

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440
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448

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