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1 **Utilization of Catalyzed Waste Vegetable Oil as a Binder for the Production of**  
2 **Environmentally friendly Roofing Tiles**

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11 **ABSTRACT**

12 Climate change has become a major issue in recent years owing to the emission of greenhouse  
13 gases. Mitigation measures are required to overcome the challenges pertained to greenhouse gases  
14 emissions. This research paper attributes to the utilization of catalyzed waste vegetable oil as a  
15 binder for the production of roofing tiles to replace the conventional construction materials such  
16 as clay and cement. A novel methodology of utilizing catalyzed waste oil incorporated with sand  
17 and filler was adopted and the innovative product produced entitled as catalyzed Vege-Roofing  
18 tiles was produced that discovered to be economical and environmentally friendly in contrast to  
19 the traditional binders. It is believed that an extended heat curing of vegetable oil results in a  
20 complex oxy-polymerization reaction converting it into a rigid binder. Triplicate prototypes

21 samples were manufactured to optimize the final conditions for the fabrication of catalyzed Vege-  
22 Roofing tiles. Optimized conditions were then implemented to produce standard catalyzed Vege-  
23 Roofing tiles and these fabricated tiles have shown flexural stress of up to 12 MPa for 18 hours of  
24 curing. Moreover, these novel tiles were tested for permeability and water absorption according to  
25 the ASTM standards and have shown impermeability and remarkably low water absorption.  
26 Progressively, the embodied energy and embodied carbon requirements for these tiles found to be  
27 0.64 MJ/kg and 0.327 kg CO<sub>2</sub> per equivalent respectively which is quite less in comparison to the  
28 traditional binders. Conclusively, environmentally friendly and economic production of tiles,  
29 conservation of existing resources and overcoming the issue of waste management are the  
30 remarkable outcomes of this research.

31 **Keywords:** Catalyzed Vege-Roofing Tile; Catalyst; CO<sub>2</sub> Emission; Embodied Carbon; Embodied  
32 Energy; Waste Vegetable Oil

### 33 **1. Introduction**

34 Owing to the concern of the effects of greenhouse gases (GHGs) on our environment, it is essential  
35 to discover solutions of mitigating these gases to secure the future (Oludolapo and Charles, 2017).  
36 Surprisingly, only building industry accounts about 40% of energy utilized globally and moreover,  
37 approximately 46% of this quantity being consumed in developing countries (Hameed, 2009). Also  
38 in the course of activities involved in construction such as extraction and enormous consumption  
39 of raw materials, a huge of amount of waste is generated (Bogas et al., 2015; Angel et al., 2017).  
40 According to International Energy Agency (IEA), approximately 10% of current global man-made  
41 CO<sub>2</sub> emissions originated from the cement industry only (Shen et al., 2016). Furthermore, 1.84 –  
42 2.8 kJ/kg of energy and a temperature higher than 1000 °C is required in the manufacturing of clay

43 tiles (Froth and Shaw, 2013). An enormous amount of carbon dioxide released during the  
44 production of these masonry units also believed to be responsible for the enhancement of global  
45 warming. This alarming situation requires being addressed seriously and environmentally friendly  
46 approaches are required to be implemented at both manufacturing and extraction of building  
47 materials to minimize the energy consumption.

48 Waste management is another serious issue of concern, since thousands of millions of tons of waste  
49 produced per year. It is estimated that for commercial frying only in UK and US, approximately  
50 50-90 million liters and 300 million gallons of used cooking oil is produced annually. Used  
51 cooking oil is considered to be a waste since it pollutes the water and affect the marine life when  
52 discharged into local streams (Forth and Zoorob, 2012). Utilization of waste vegetable oil in the  
53 production of Biodiesel has already paved a way to overcome the issue of waste oil disposal, but  
54 escalating cost of production and by-product disposal are the major hurdles in the implementation  
55 of this process (Forth and Zoorob, 2012; Nadyaini et al., 2011). Similarly, fly ash that is available  
56 in huge amount from thermal power plant and considered as a hazardous waste is usually land  
57 filled. It is revealed that when fly ash content between 30 to 60% is used, a concrete having good  
58 mechanical strength and durability could be produced (Marceau, 2002). It is also reported that a  
59 20% fly ash as filler has higher compressive strength as compared to more than 50% fly ash as  
60 filler (Haiying et al., 2007). Another notable consideration is that high percentage of filler could  
61 have a negative impact on the compressive strength of building material (Naik et al., 2004).  
62 Moreover, utilizing fly ash content greater than 50% could reduce the workability of the mixture  
63 and also induce difficulty to compact the mixture (Garbacz and Sokołowska, 2013).  
64 Till today, more stress is laid on investigating the replacement of the aggregates with waste  
65 materials but unfortunately, a very limited focus has been paid in evaluating the energy

66 requirements of binders like cement production or kiln firing during production of concrete and  
67 clay tile respectively. Cement production and firing clay bricks are considered as the foremost  
68 contributors in increasing the embodied energy and CO<sub>2</sub> requirements (Jones and Hammond,  
69 2008). Contrary, aggregate has negligible impact on the energy emissions and consequently on the  
70 environment. Substituting the masonry units with renewable materials like vegetable oil having  
71 comparatively lower energy and carbon emissions will definitely contribute much to overcome the  
72 threatening issue of global warming. However, the concept of utilizing vegetable oil as an effective  
73 building material is limited to few studies only. Oxy-polymerization reaction is considered as  
74 responsible for the binding effect since it increases the viscosity and consequently hardening of  
75 the vegetable oil (Quesnel, 1994; Johnson et al., 2015). It is investigated that building blocks could  
76 be produced from vegetable oil mixed with recycling aggregates. These blocks have shown the  
77 potential to replace the conventional building blocks (Forth and Zoorob, 2012). In addition,  
78 building blocks produced by the encapsulation of vegetable oil and petroleum sludge have shown  
79 high compressive strength compared to traditional building blocks (Johnson et al., 2015). It is also  
80 revealed that virgin vegetable oil mixed with aggregate and filler followed by compaction and heat  
81 curing could be utilized for the production of roofing tiles (Noor et al., 2015). However, energy  
82 and economic constraints have limited the feasibility of these processes. Hung et al. (2015)  
83 reported that a blend of waste vegetable oil and glycerol can be used in the production of masonry  
84 units. The blocks produced have shown high compressive strength and low energy emissions in  
85 contrast to concrete blocks. However, changes in EU Directive restricted the generation of bio-  
86 fuel and consequently the glycerol's production and thus limited the feasibility of the process. It  
87 was hypothesized in the present innovation that sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) added as a catalyst to waste  
88 vegetable oil would reduce the curing time and assist in developing a more energy efficient and

89 economical process. The present investigation aims to develop an alternate binder to replace the  
90 conventional environmentally unfriendly binders for the production of masonry units. Catalyzed  
91 Vege-Roofing tiles produced by incorporation of catalyzed waste vegetable oil and filler and sand  
92 were examined for flexural strength, water absorption, and permeability according to ASTM  
93 standards. Environmental aspects were also determined by calculating the embodied energy and  
94 carbon emissions. Furthermore, cost was also calculated for catalyzed Vege-Roofing tiles and  
95 comparative analysis was carried out with conventional concrete roofing tiles. Induction of this  
96 novel binder for the production of masonry units would expect to reduce the energy emissions and  
97 cost of the building sector to an enormous extent.

## 98 **2. Experimental procedure**

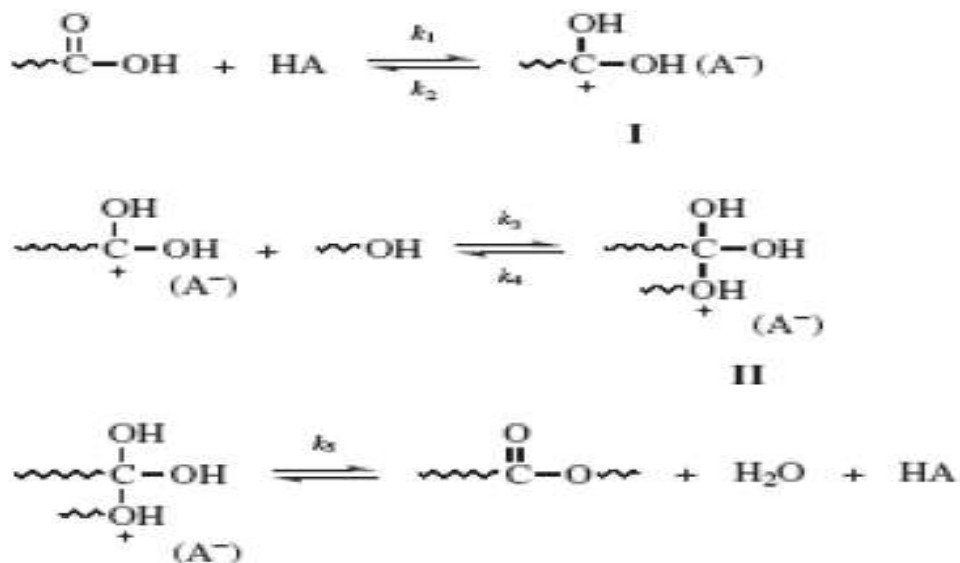
### 99 *2.1 Materials*

#### 100 *2.1.1 Binder*

101 Catalyzed vegetable oil constitutes of waste vegetable oil and  $H_2SO_4$ , was used as a binder in this  
102 investigation. Waste vegetable oil was obtained from the local restaurants in Sri Iskandar,  
103 Malaysia. Some of the properties of the waste vegetable oil examined are listed in Table 1.  
104 Moreover, 8.33 M concentrated sulfuric acid ( $H_2SO_4$ ) with a percentage purity of 96 to 98% and  
105 brand name Qrec was utilized as catalyst with waste vegetable oil to reduce the curing time in the  
106 production of roofing tiles. The ratio of waste oil to  $H_2SO_4$  used before attaining the optimized  
107 value was 25:1.

108 Waste vegetable oil consists of a mixture of fatty acids and mono, di- and tri-glycerides. Upon  
109 prolonged heating, some of the fatty acids may form dimers or trimers with more than one  
110 carboxylic acid group. Hence, poly-esterification can occur between di-acids and diols found in  
111 waste vegetable oil to form solid polyester and bind the tile material effectively during its curing

112 process. Esterification process is catalyzed by Bronsted acids, preferably by sulfonic and sulfuric  
 113 acids. These catalysts give very high yields in alkyl esters (Schuchardta et al., 1998). The  
 114 mechanism of the acid-catalyzed esterification between a carboxylic acid and an alcohol is shown  
 115 in Fig. 1. The protonation of the carbonyl group of an acid leads to the carbonation I which, after  
 116 a nucleophilic attack of the alcohol, produces the tetrahedral intermediate II, which eliminates a  
 117 water molecule to form the ester and to regenerate the catalyst HA. Esterification is an equilibrium  
 118 reaction and the transformation occurs essentially by mixing the reactants. However, the presence  
 119 of a catalyst (typically a strong acid) accelerates considerably the rate of reaction.



120  
 121 **Fig.1:** Mechanism of the acid-catalyzed esterification between a carboxylic acid and an alcohol

122 **Table 1:** Some tests on waste vegetable oil

Property	Used vegetable oil collected	Maximum standard limit (Berger, 2005)
Acid Value (%)	3.7	2.5
Free Fatty acids (%)	3.5	2.5

Oxidized fatty acids (%)	1	2.1
Total polar molecules (%)	30	25-27

123

124 *2.1.2. Sand Aggregate*

125 Two types of sand named as river and mining sand is used in this process. Specific gravity of both  
 126 types of sands is determined by helium ultra-pycnometer and large pycnometer method following  
 127 the ASTM C127-88 and C128-88. In addition, size distribution attained by sieving analysis of river  
 128 and mining sand is conducted according to ASTM C 136. Size gradations of river and mining  
 129 sands are presented in Fig. 2.

130

131 **Table 2:** Specific Gravity of Sand Types

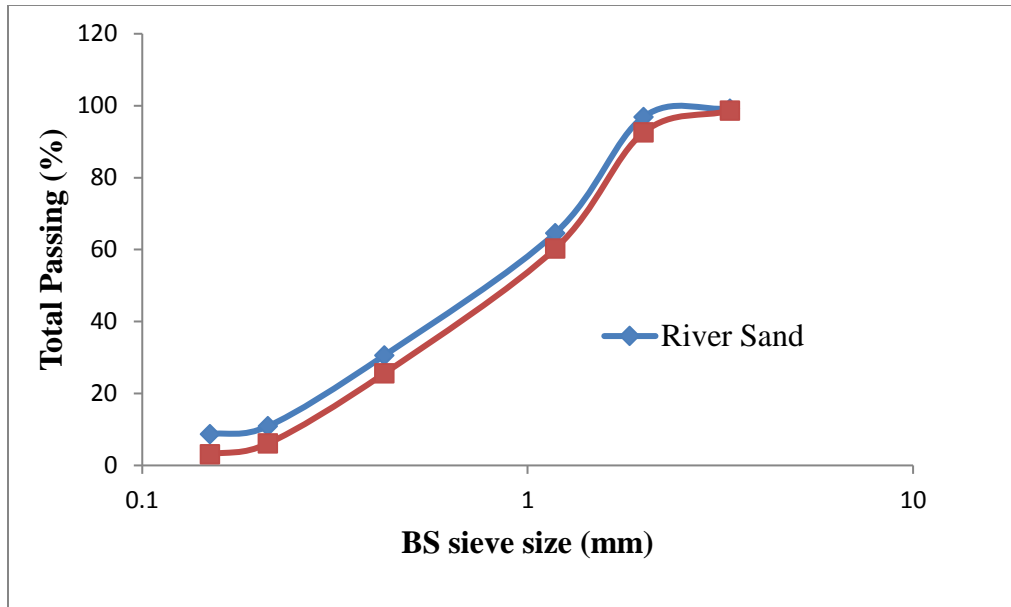
Type	Helium Pycnometer	ASTM C127
River Sand	2.53	2.570
Mining Sand	2.67	2.647

132

133

134





135

136 **Fig. 2:** Size gradation of the sand aggregates

137 *2.1.3. Filler*

138 Fly ash, having class F, size less than 75  $\mu\text{m}$  and specific gravity of 2.5 was purchased from Kah  
 139 Hwa Industries SDN. BHD, Malaysia. The typical chemical compositions and oxide analysis for  
 140 fly ash used in this investigation are shown in Table 3. Fly ash was utilized as filler in the  
 141 production of both prototypes samples and catalyzed Vege-Roofing tiles.

142 Table 3: Chemical Composition of Fly ash

Components	Mass (%)	Standard Limits (ASTM C618) %
Silicon oxide ( $\text{SiO}_2$ )	60.52	$\text{SiO}_2$ plus $\text{Al}_2\text{O}_3$ plus
Aluminium oxide ( $\text{Al}_2\text{O}_3$ )	31.12	$\text{Fe}_2\text{O}_3$ , min 70%
Ferrous oxide ( $\text{Fe}_2\text{O}_3$ )	1.46	Obtained 93.1%

Calcium oxide (CaO)	3.81	
Sodium oxide (Na <sub>2</sub> O)	1.21	
Magnesium oxide (MgO)	0.84	
Sulfur trioxide (SO <sub>3</sub> )	0.73	Max 5
Chloride as Cl	0.06	
Loss of Ignition (LOI)	0.86	Max 6

143

144 *2.2. Methodology*

145 Primarily, parameters were required to be optimized before the fabrication of catalyzed Vege-  
146 Roofing tiles. Parameters that needed to be optimized are fly ash content, curing time, the amount  
147 of catalyst, blending time of catalyzed waste vegetable oil and storage life of catalyzed waste oil.  
148 The temperature evaluated as appropriate for production of all samples was 190°C (Noor et al.,  
149 2015). Initial appropriate values used for the optimization process of each parameter are displayed  
150 in Table 4. After scrupulous mixing (catalyzed oil did not cause stickiness and gulping with  
151 aggregate and filler based on physical observation) of waste oil and catalyst with aggregate and  
152 filler, the mixture was transported to standard Marshall Moulds (50 mm × 100 mm) and compacted  
153 with 10 blows. Triplicate prototypes samples were then heat cured in an oven maintained at a  
154 temperature of 190°C and tested for flexural stress to attain the optimized values for each  
155 parameter. Highest flexural stress developed was considered as a criterion for the optimization of  
156 each parameter.

157

158

159 **Table 4: Initially selected values of parameters for the optimization process**

Parameter	Selected Value
Filler	35% of total (sand + fly ash) (Noor et al., 2015)
Temperature	190°C (Noor et al., 2015)
Waste oil content	8% of total (sand + fly ash). Based on physical observation, i.e it do not cause stickiness and gulping with aggregate and filler.
Acid content	6% of total waste oil used or waste oil to acid ratio of 25:1 (Based on physical observation of change of waste oil’s color to dark brown)
Catalyzed Waste oil Content	Catalyzed Waste oil / Aggregate (sand) + Filler (fly ash) = 0.0945 10 minutes blending time of waste oil and catalyst Fresh catalyzed waste oil

160

161 Notably, similar ratio was chosen for catalyzed waste oil to other materials (sand and fly ash) for  
 162 the production of all samples to that used for water to other materials (cement and sand) for the  
 163 production of concrete roofing tiles (Johansson, 1995). However, catalyzed waste oil comprised

164 of both catalyst and waste oil, optimized value for the percentage of catalyst in waste oil was also  
165 needed to be calculated. Density, specific gravity, and porosity of optimized prototypes were then  
166 determined for the optimized prototypes and further tested for a percentage of water absorption  
167 and permeability respectively compiled with the ASTM standards.

168 After achieving the optimized values for each parameter, final catalyzed Vege-Roofing standard  
169 tiles of dimensions 390 mm x 240 mm x 10 mm were produced (Johansson, 1995). Catalyzed  
170 Vege-Roofing tiles were then examined for flexural stress, percentage of water absorbed and  
171 permeability respectively (ASTM C67-13; ASTM C 1167-03; ASTM C 1492-03; WSDOT 802).  
172 In addition, energy emissions and economic characteristics of these novel roofing tiles were also  
173 evaluated. Energy characteristics of roofing tiles were demonstrated by calculating the embodied  
174 energy and embodied carbon. Embodied energy was calculated by multiplying the amount of each  
175 material required in producing a single catalyzed Vege-Roofing tile with the embodied energy  
176 requirements of that particular material. The determination of total carbon emissions were carried  
177 out by the use of life cycle assessment (LCA) method. Environmental impact was determined for  
178 different stages of catalyzed roofing tiles such as cradle to gate, manufacturing, distribution and  
179 end of life. Carbon emission factors for processes and materials were attained by using ecoinvent  
180 3.3. Total carbon emissions for catalyzed Vege-Roofing tiles were then assessed in accordance to  
181 LinkCycle Quick LCA tool. Assumptions that were used in calculating the embodied energy and  
182 embodied carbon are enlisted in Table 5. Cost was determined based on the raw materials and  
183 utility charges per tile and compared with the conventional concrete roofing tile. Other  
184 miscellaneous charges were excluded from the calculations. Manufacturing steps for standard  
185 catalyzed Vege- Roofing tiles are displayed in Fig. 3.

186

187 **Table 5:** Assumptions in calculating energy emissions

Process	Assumption
Transportation	1. Mode of Transportation was lorry 16-32 metric ton, EURO6.
	2. Waste vegetable oil was collected from local restaurants up to a distance of 1000 Km.
Production	1. Large oven having a capacity of 15 KWh was used for the production of tiles.
	2. Approximately 900 tiles were fabricated in an oven in a single batch.
Distribution	1. Roofing tiles were distributed up to a distance of 200 Km
End of Life Management	1. Used Roofing tiles were disposed of in a local site at a distance of 40 Km.

188

189 *2.3. Testing Procedures*

190 *2.3.1. Density and Porosity*

191 Density, specific gravity and porosity were determined for all the samples to ensure uniformity in  
 192 test results by using equations (1) and (2) and (3).

193 
$$S.G_{mix} = (m_{filler} + m_{sand} + m_{oil} + m_{acid}) / (m_{filler}/S.G_{filler} + m_{sand}/S.G_{sand} + m_{oil}/S.G_{oil}$$
  
 194 
$$+ m_{acid}/S.G_{acid} ) \quad (1)$$

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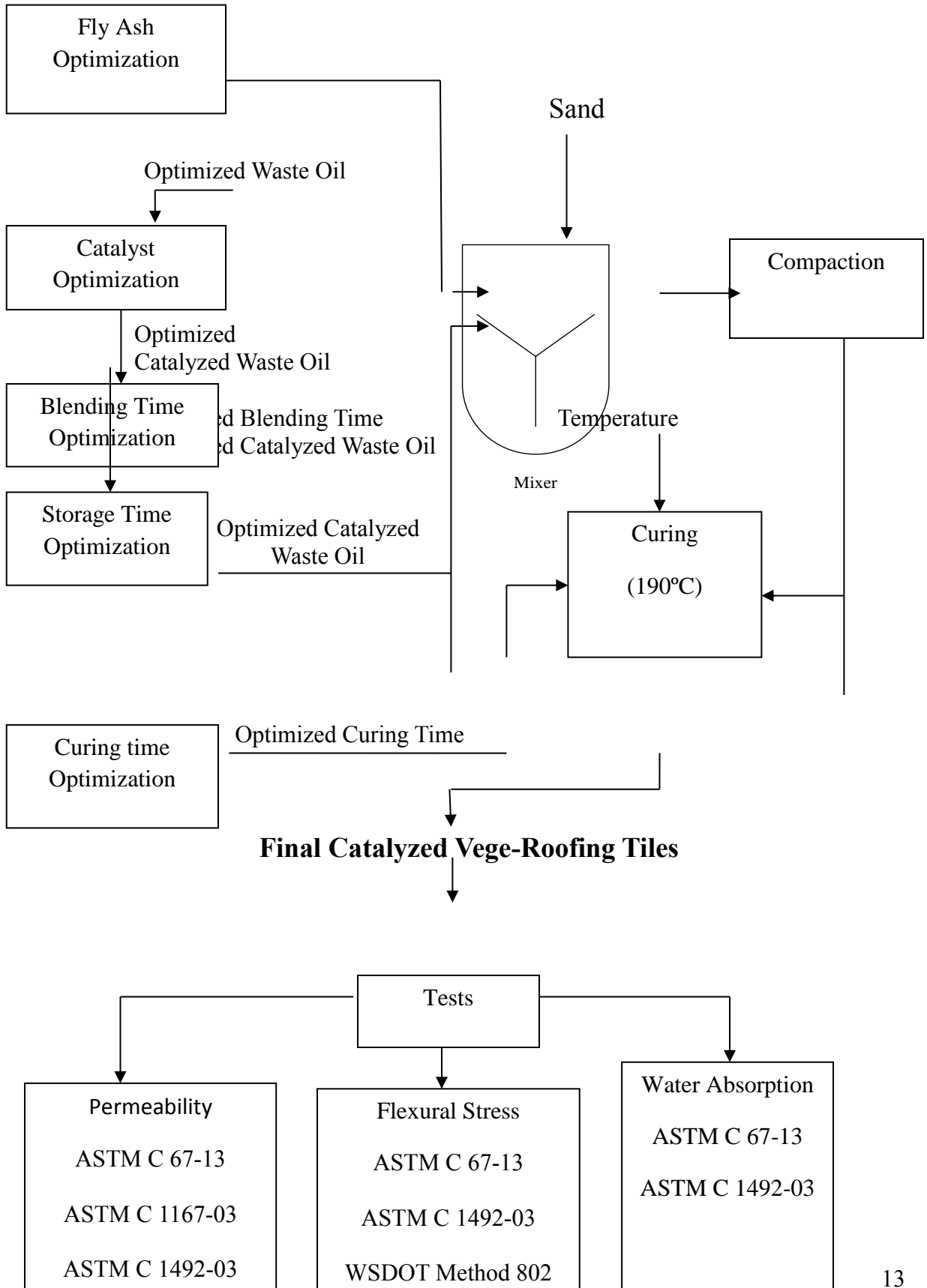
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213

214 **Fig. 3:** Catalyzed Vege-Roofing tile manufacturing steps

215



216

217 **Fig.4:** Catalyzed Vege-Roofing tile sample

218 
$$P=100 (1-D_{mean} / (S.G)) \quad (2)$$

219 
$$D= M/V \quad (3)$$

220 Density was also measured by a standard method (WSDOT TM 810) to demonstrate the validity  
221 of the results.

222 
$$D = A \times d_w / A-B \quad (4)$$

223 where,

224  $S.G$  =specific gravity (unit less)

225  $D$ = density in  $g/cm^3$

226  $P$ = Porosity in %

227  $A$ = Mass in grams of the surface dry sample in air

228  $B$ = Mass in grams of the sample in water

229  $d_w$  = density of the water at test temperature

### 230 2.3.2. *Water Absorption*

231 To evaluate the quantity of water that a brick can absorb, water absorption test was conducted  
232 according to standard methods (ASTM C 67-13; ASTM C 1492-03).

233 The water absorption was calculated as:

$$234 \text{ Absorption } (C), \% = 100 (W_s - W_d) / W_s \quad (5)$$

235 where,

236  $W_d$  = Mass in air in g

237  $W_s$  = Saturated mass in water after 24 hours in g

238 Boiling water absorption can be determined as

$$239 \text{ Boiling Absorption, } B \% = 100 (W_b - W_d) / W_b \quad (6)$$

240 Finally, the saturation coefficient can be calculated as:

$$241 \text{ Saturation Coefficient} = C/B \quad (7)$$

### 242 2.3.3. *Permeability*



243 Permeability, one of the unwanted features for roofing tiles was determined for prototypes samples  
244 and standard catalyzed roofing tiles according to ASTM standard method (ASTM C 67-13; ASTM  
245 C 116703; ASTM C 1492-03). Tile's samples placed on stand in such a way that its undersides  
246 were visible and water up to approximately 10mm height allowed to sit on the top of the samples  
247 for 24 hours. The area between the samples and setup was properly sealed with a sealant to prevent  
248 the leakage. The water drops were inspected after 24 hours and if more than two of them found on  
249 the underside of the samples, then the samples would consider as significantly permeable. The  
250 experimental setup of permeability test for prototypes samples and standard tiles is shown in Fig.  
251 5.



252

253 **Fig. 5:** Permeability setup for a) Prototypes samples b) Standard tiles

#### 254 2.3.4. Flexural Strength

255 Flexural strength indicates the load that a material can withstand without breaking or rupture.  
256 Flexural strength for prototypes samples was determined by ASTM standard methods (ASTM C  
257 67-13; ASTM C 1492-03) denoted by  $\sigma$  and is expressed in MPa.

258  $\sigma = MC/I$  (8)

259 where,

260 M = unit load in Newton

261 C = distance from the neutral axis in millimeters

262 I = moment of inertia in millimeters

263 Moreover, flexural stress of final roofing tiles was determined in accordance with three points  
264 bending test (ASTM C 67-13; ASTM C 1492-03; WSDOT 802)

265  $Flexural\ stress = 3*P*L / 2*W*d^2$  (9)

266 where,

267 P =Loading force in Newton

268 L = Span Length of the tile in millimeters

269 W = Width of the tile in millimeters

270 d = Thickness of the tile in millimeters

### 271 **3. Results and discussions**

#### 272 *3.1 Optimizations*

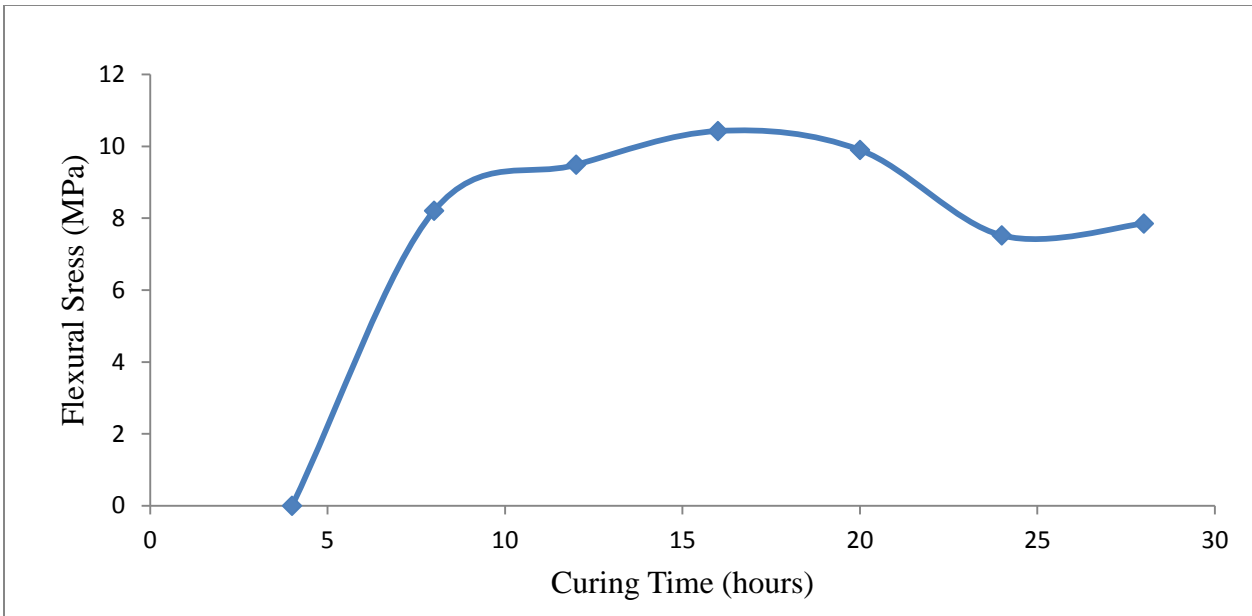
##### 273 *3.1.1 Curing Time*

274 Triplicate prototypes specimens were produced at initially suitable conditions (35% filler, 65%  
275 sand, 8% of waste oil content to aggregate and filler, waste oil to H<sub>2</sub>SO<sub>4</sub> ratio of 25:1, 10 minutes

276 blending of catalyzed waste oil) and cured for different hours at a temperature of 190°C. Fig. 6  
277 presents the trend of flexural stress developed for varying curing time at a temperature of 190°C.  
278 It is revealed from Fig. 6 that the flexural stress discovered to be highest for 16 and 20 hours of  
279 curing. Moreover, after 20 hours of curing at a temperature of 190°C, flexural stress began to  
280 decline to an appreciable extent probably due to the reason that prolonged curing induced the  
281 cracks internally causing the strength of prototypes specimens to reduce. It is also discovered from  
282 Fig.6 that flexural stress achieved for all days of curing has fulfilled the standard minimum  
283 requirement of 6 MPa (Crow, 2000). Moreover, standard deviation for each value of flexural stress  
284 found to be less than 0.01 thus indicating a well-controlled process (NRMCA, 2000). **Efficient**  
285 **curing of the tiles is extremely important since insufficient curing can reduce the strength to a**  
286 **tremendous extent. To ensure the adequate curing of the final tiles and taking into account the**  
287 **energy perspective**, the average of two curing times that showed highest flexural stress i.e. 18  
288 hours of curing considered as an optimal curing time for the production.

### 289 *3.1.2 Filler Content*

290 To acquire suitable filler percentage, triplicate prototype specimens were produced at initially  
291 suitable conditions of catalyzed waste oil and optimized curing time of 18 hours and filler  
292 percentages were varied between 30% and 50%. Filler content was restrained between 30 and 50%  
293 since utilizing fly ash content greater than 50% could reduce the workability of the mixture and  
294 also induce difficulty to compact the mixture (Garbacz and Sokołowska, 2013). Flexural stress of  
295 triplicate prototype specimens at varying filler percentages was determined and illustrated in Fig.  
296 7.



297

298 **Fig. 6:** Optimization of curing time

299 The trend of Fig. 7 reveals that flexural stress was on the lower side with 30% fly ash as filler. By  
 300 increasing the fly ash content to 35%, highest flexural stress of approximately 9 MPa was achieved.

301 However, further addition of fly ash reduced the flexural stress for the samples as observed for  
 302 filler percentages of 40%, 45% and 50% respectively. However, no specific mechanism is

303 available for the reaction of vegetable oil and fly ash but it is observed that fly ash improves the  
 304 binding characteristics of vegetable oil and enhances the flexural and tensile properties due to the

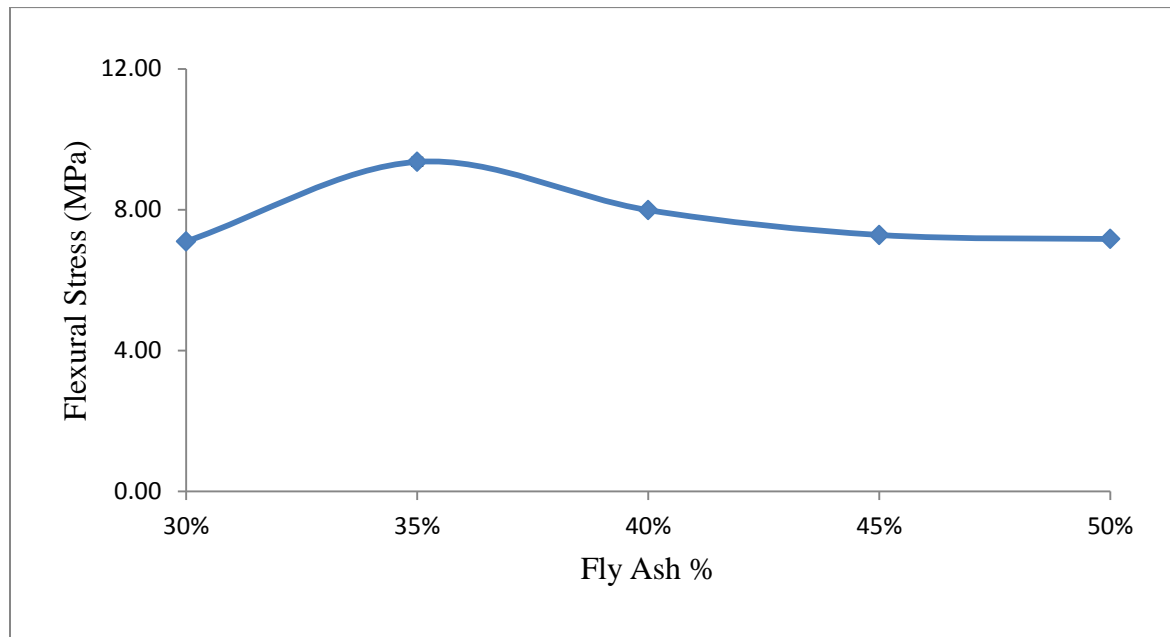
305 presence of high silica and alumina content (Micheal et al., 2014; Saumya et al., 2016). Moreover,  
 306 fly ash has a neutralizing effect in strong acids such as sulfuric acid i.e. when introduced into

307 strong acids it tends to lower the pH (Manisha et al., 2009). This is a remarkable effect since it is  
 308 an indication that catalyzed Vege-Roofing tiles when exposed to fire would be non-flammable.

309 Nonetheless, the flexural stress developed and standard deviations for each filler percentage were  
 310 higher in contrast to standard minimum requirement (CROW, 2000; NRMCA, 2000). It can be

311 deduced from Fig. 7 that 35% of fly ash as filler showed highest flexural stress and considered as  
312 the optimal percentage of filler content.

313



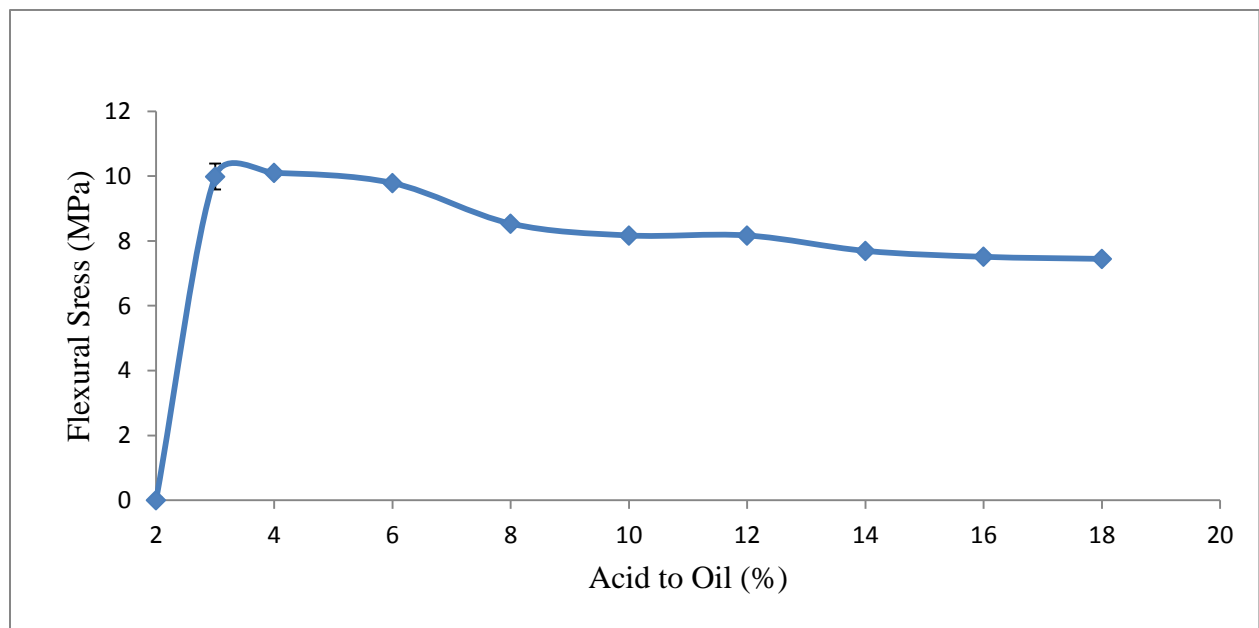
314

315 **Fig. 7:** Optimization of filler content

### 316 *3.1.3 Percentage of Acid*

317 Triplicate prototypes samples were produced at initially selected conditions of blending and  
318 storage time and optimized fly ash content of 35% and optimized curing time of 18 hours. Acid  
319 percentage to waste oil is altered in the range of 3% to 18 %. The range was chosen based on  
320 physical observation since introducing 3% of acid into waste oil just started the color change of  
321 catalyzed waste oil blend. Moreover, increasing percentage of an acid in oil beyond 18% reduced  
322 the workability of the mixture since the addition of an acid into waste oil tends to increase the  
323 viscosity the mixture of acid and oil. Fig. 8 exhibits the flexural stress achieved with varying  
324 percentage of acid to waste oil. It is demonstrated from Fig. 8 that flexural stress found to be

325 highest between 3 to 6 percent of acid in waste oil. It is also discovered that increasing the  
326 percentage of acid in waste oil reduced the flexural stress of samples. This is probably due to the  
327 increased viscosity of the catalyzed mixture when additional acid added into waste oil. Oxy-  
328 polymerization reaction is considered responsible for increased viscosity of the oil (Quesnel, 1994;  
329 Johnson et al., 2015). It is revealed that upon prolonged heating, some of the fatty acids in waste  
330 vegetable may form dimers or trimers with more than one carboxylic acid group. Hence, poly-  
331 esterification can occur between di-acids and diols found in waste vegetable oil to form solid  
332 polyester and bind the tile material effectively during its curing process at 190°C. It is also believed  
333 the presence of a catalyst (typically a strong acid like H<sub>2</sub>SO<sub>4</sub>) accelerates considerably the rate of  
334 reaction and effectively reduced the curing time to about 18 hours (Schuchardta et al., 1998). The  
335 flexural stress and standard deviation achieved for each acid percentage calculated to be 8 to 10  
336 MPa and 0.001 to 0.4 MPa respectively which is well within the practical standard limits (CROW,  
337 2000; NRMCA, 2000). Highest flexural stress was achieved for 3 to 4 percent of acid in waste oil  
338 and was utilized for fabrication of standard catalyzed Vege-Roofing tiles.



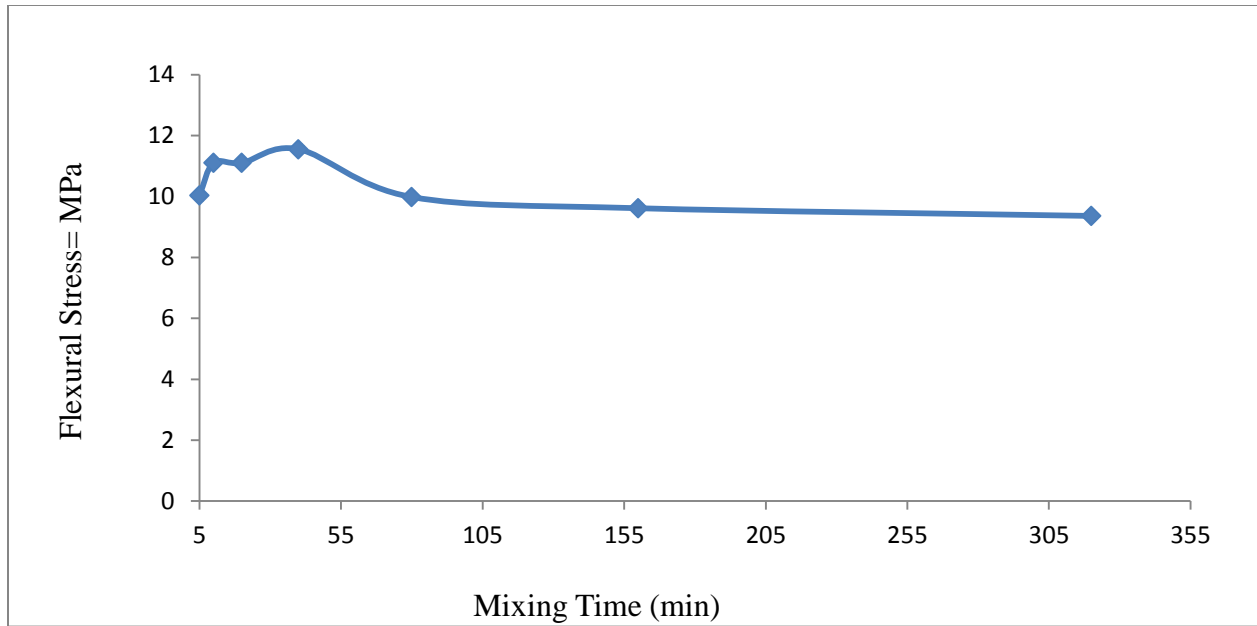
339

340 **Fig. 8:** Optimization of Acid content

341

#### 342 *3.1.4. Blending Time for Catalyzed Waste Oil*

343 Blending or mixing time of catalyzed waste oil (mixture of acid and waste oil) is considered as  
344 one of the significant parameters to be reported since proper mixing of waste oil and acid could  
345 improve the mechanical properties of the catalyzed Vege-Roofing tiles. Waste oil and acid were  
346 blended for different times and triplicate prototypes samples were produced from each blend  
347 utilized as fresh at optimized conditions of filler, curing time and percentage of acid in waste oil.  
348 Blending time for catalyzed waste oil was then optimized by demonstrating the flexural stress of  
349 triplicate prototypes samples produced. Fig. 9 presents the flexural stress achieved by prototypes  
350 samples produced from various blending times of optimized catalyzed waste oil at 190°C. It is  
351 illustrated by Fig. 9, that flexural stress developed found to be highest for blending times of 10, 20  
352 and 40 minutes respectively. Interestingly, after 40 minutes of blending the flexural stress did not  
353 change much probably due the reason that optimum mixing has been achieved earlier. However,  
354 from energy perspective, 10 minutes of blending was evaluated as an optimal blending time for  
355 the production of standard catalyzed Vege-Roofing tiles.



356

357 **Fig.9:** Optimization of blending time of catalyzed waste oil

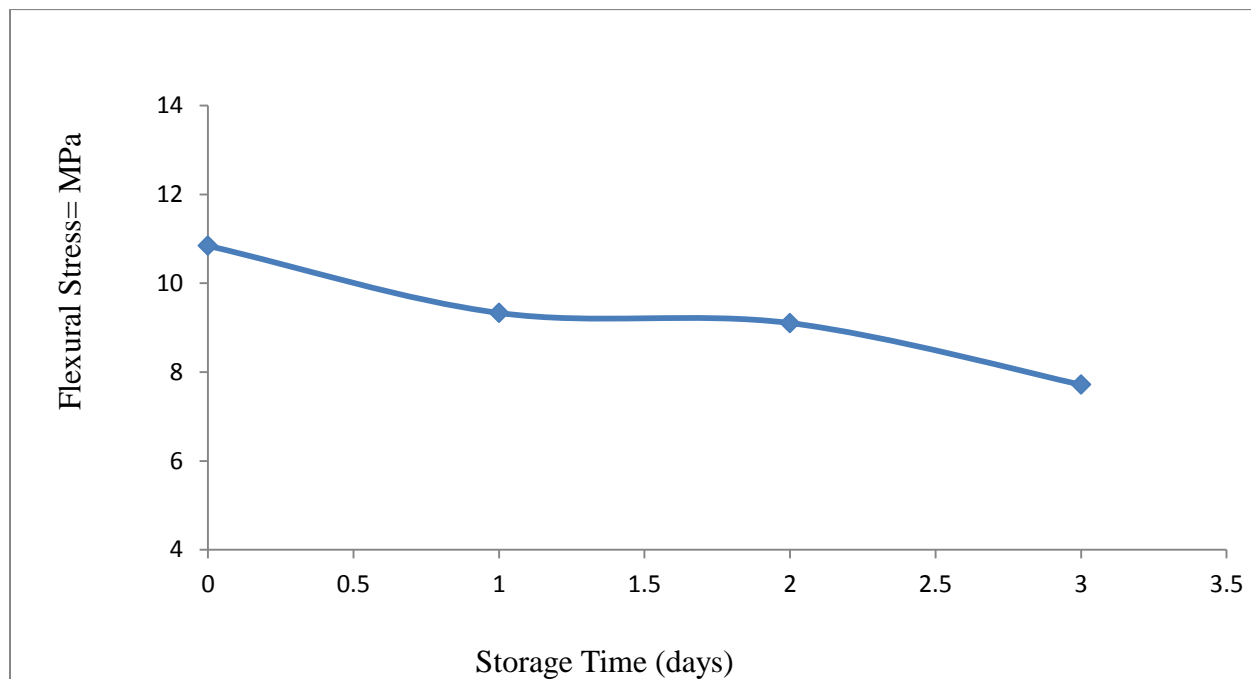
358 *3.1.5 Storage Life for Catalyzed Waste Oil*

359 Storage life is an indication of storing the material while remaining within a safe limit. Storage life  
 360 consideration of catalyzed waste oil is important especially from industrial point of view. Flexural  
 361 stress for triplicate prototypes samples produced from fresh and stored catalyzed waste oil at  
 362 already optimized conditions is displayed in Fig. 10. Notably, after 3 days of storage, catalyzed  
 363 waste oil evaluated as no longer workable with the aggregate and filler since it converted to a very  
 364 rigid material. It is revealed from the Fig. 8 that flexural stress found to be on the higher side for  
 365 fresh catalyzed waste oil and started to decline with days of storage. It is shown in Fig.10 that  
 366 utilizing the freshly catalyzed waste oil attained a flexural stress of approximately 11 MPa which  
 367 is quite high in contrast to the standard minimum requirement of 6 MPa (CROW, 2000).  
 368 Additionally, the standard deviation of flexural stress for each day of storage of catalyzed waste



369 oil found to be less than 0.1 MPa. Thus freshly catalyzed waste vegetable oil was evaluated as the  
370 optimal value for the fabrication of standard catalyzed Vege-Roofing tiles.

371



372

373 **Fig. 10:** Storage life optimization of catalyzed waste Oil

### 374 3.2 Optimized Prototypes

375 Prototypes samples were produced with already optimized parameter and examined before the  
376 fabrication of standard catalyzed Vege-Roofing tiles. **Density, porosity, permeability and**  
377 **percentage of water absorption were calculated for optimized prototypes samples.** Table 6 shows  
378 that high density specimens indicated low porosity while specimens that had low density pointed  
379 relatively high porosity. **This is an indication that a dense material has fewer chances to leak or**  
380 **have low porosity.** Results of Table 6 also reveal that percentage of water absorption for prototypes  
381 samples produced was in the range of 1.8% to 2.6% which is quite less in contrast to the standard

382 practical limit (Johansson, 1995; ASTM C 1492-03). Moreover, it should also be taken into  
 383 account that all the optimized prototypes samples have passed the permeability test. The low  
 384 percentage of water absorption is a reflection of the low porosity of the tiles (Hung et al., 2015;  
 385 Zhang and Zong, 2014). Moreover, higher the porosity more will be the probability that the  
 386 material becomes permeable (Ekstorm, 2001). Generally, greater the permeability, lower will be  
 387 the durability performance of concrete (Khan and Lynsdale, 2002). Less porosity, low percentage  
 388 of water absorption and impermeability enhance the durability of the tiles (Farhana et al., 2015).

389 **Table 6: Density and porosity of optimized prototypes specimens** (35% filler, 65% sand, 8% oil,  
 390 0.5% acid,  $S.G_{\text{filler}}=2.5$ ,  $S.G_{\text{sand}}=2.66$ ,  $S.G_{\text{oil}} = 0.84$ ,  $S.G_{\text{acid}}=1.83$ ,  $S.G_{\text{mix}}=2.1$ )

391

No.	Mass (g)	Density $\text{g}/\text{cm}^3$	Porosity (%)	Water absorption (%)	Permeability Test
1	164.0	2.08	1.0	2.12	PASS
2	162.5	2.06	2.0	2.45	PASS
3	163.5	2.07	1.5	1.98	PASS
4	162.1	2.05	2.4	2.76	PASS
5	160.9	2.04	2.9	3.16	PASS

392

393

394

395 *3.3 Standard Catalyzed Vege-Roofing tiles*

396 Optimized prototypes samples found to be impermeable and showed a low percentage of water  
 397 absorption. Thus, the utilization of catalyzed vegetable oil was further investigated by producing  
 398 standard catalyzed Vege-Roofing tiles at already optimized conditions at a temperature of 190°C.  
 399 Initially, density and porosity of standard catalyzed Vege-Roofing tiles were calculated and  
 400 indicated in Table 7. The bulk density was approximately 1.98 to 2 g/cm<sup>3</sup> while the density varies  
 401 from 1.7 to 2.4 g/cm<sup>3</sup> for lightweight to normal concrete (Richard, 2004). It is found that denser  
 402 materials have fewer chances to leak (Dias, 2000). It is also revealed from the Table 7 that denser  
 403 materials had low porosity and vice versa (Dias, 2000 ; Ekstorm, 2001).

404 **Table 7: Density and porosity of standard catalyzed Vege-Roofing tiles cured for 18 hours** (35%  
 405 filler, 65% sand, 8% oil, 0.5% acid, S.G<sub>filler</sub>=2.5, S.G<sub>sand</sub>=2.66, S.G<sub>acid</sub>=1.83, S.G<sub>mix</sub>=2.1)

No.	Mass (g)	Density, M/V g/cm <sup>3</sup>	Density WSDOT TM 810 g/cm <sup>3</sup>	Density (Mean)	Porosity (%)
1	1870	1.99	1.97	1.98	5.8
2	1882	2.00	1.99	1.99	5.3
3	1904	2.03	2.00	2.01	4.3
4	1849	1.97	1.98	1.98	5.8
5	1858	1.98	2.00	1.99	5.3

406

407 *3.3.1 Flexural Stress & Breaking Strength*

408 Flexural stress and breaking strength calculated for catalyzed Vege-Roofing tiles produced at  
409 optimized parameters are displayed in Table 8. It can be seen from Table 8 that flexural stress  
410 developed for standard catalyzed Vege-Roofing tiles found to be in the range of 11.4 to 12.2 MPa  
411 respectively. This suggests that the average flexural stress achieved by catalyzed Vege-Roofing  
412 tiles was approximately twice in contrast to the minimum standard requirements of 6 MPa  
413 (CROW, 2000). Nonetheless, the values of flexural stress obtained were much higher as compared  
414 to the BS 6073. This is due to an excellent binding ability of waste vegetable oil incorporated with  
415 fly ash and sand. Heat curing of waste vegetable oil initiated the oxy-polymerization reaction and  
416 converted it into a solid rigid binder (Johnson, 2015; Quesnel, 2009). Thus, a high flexural stress  
417 was developed with waste vegetable oil in comparison to other wastes such as cotton and limestone  
418 powder wastes that achieved a maximum flexural stress up to 3.5 MPa (Halil and Tugut, 2007).  
419 Also, breaking strength for concrete roofing tiles should be at least 550 to 600 N and contrary,  
420 breaking strength achieved by catalyzed Vege-Roofing tiles was higher in comparison to minimum  
421 breaking strength requirements of concrete roofing tiles (Wood and Hack, 1986; BS EN, 2011).  
422 Moreover, the standard deviation for the five tiles tested for flexural stress was below 0.7 MPa  
423 indicating a well controlled process (NRMCA, 2000). This suggests that catalyzed Vege-Roofing  
424 tiles have fulfilled the minimum standard requirements of flexural stress for the production of  
425 roofing tiles.

426 **Table 8:** Breaking strength and flexural stress of optimized standard catalyzed Vege-Roofing  
427 tiles cured for 18 hours at 190°C (Span Length=130mm, Width of tiles=240mm, Depth of  
428 tiles=10mm)

Tile No.	Loading force, P (N)	Breaking Strength (N)	Flexural Stress (MPa)
1	1400	758.3	11.4
2	1500	812.5	12.2
3	1500	812.5	12.2
4	1500	812.5	12.2
5	1400	758.3	11.4

429

### 430 3.3.2 Water Absorption

431 Percentage of water absorption for standard catalyzed Vege-Roofing tiles calculated according to  
432 standard method and the results are demonstrated in Table 9. Table 9 indicates that percentage of  
433 water absorption for five catalyzed Vege-Roofing tiles was found to be in the range of 4.7 to 5.4  
434 percent. It indicates that values of water absorption attained were within the standard limit  
435 (Johansson, 1995; ASTM C 1492-03; Donald and Grail, 1985). The low percentage of water  
436 absorption was thought to be due to the low porosity of the tiles (Hung et al., 2015; Zhang and  
437 Zong, 2014). In addition, the boiling water absorption is usually more as compared to fresh water  
438 absorption. The average boiling absorption was approximately 7.4% which is low in contrast to

439 the boiling water absorption of building blocks produced from encapsulation vegetable oil and  
440 petroleum sludge (Johnson et al., 2015). Since the percentage of water absorption under boiling is  
441 still within the standard practical limit, it indicates that catalyzed Vege-Roofing tiles can work well  
442 in extremely hot and humid conditions (ASTM C67-13; ASTM C1492-03). Boiling water  
443 absorption is usually determined to find the saturation coefficient of tiles. Saturation coefficient of  
444 catalyzed Vege-Roofing tiles found to be in the range of 0.64 to 0.78 which is within the standard  
445 limit of less than 1 (BIA, 2007). A low value of saturation coefficient is an indication that the tiles  
446 are less absorptive and more durable while tiles with high saturation coefficients are susceptible  
447 to damage (Abdullah et al., 2015). Furthermore, all the catalyzed Vege-Roofing tiles tested for  
448 permeability have passed the test. Less porous materials are denser and denser materials have  
449 fewer chances to leak and vice versa. Thus higher the density lower will be the porosity and  
450 permeability (Dias, 2000; Zhang and Zong, 2014). Low porosity, low percentage of water  
451 absorption and impermeability increases the durability of the tiles (Farhana et al., 2015).

### 452 *3.3.3 Energy Characteristics*

453 Embodied energy and embodied carbon for producing catalyzed Vege-Roofing tiles were assessed  
454 to ensure the environmental suitability of the product. Embodied energy for same batch of tiles  
455 produced per catalyzed Vege-Roofing tiles is displayed in Table 10. Since fly ash is the by-product  
456 produced during combustion of coal in electricity generation, it has no process energy. In addition,  
457 embodied energy of fly ash is zero since it is a waste and its collection is obligatory (Chani et al.,  
458 2003; Ostwal and Chitawadagi, 2014).

459

460 **Table 9:** Percentage of water absorption and saturation coefficient for optimized standard  
 461 catalyzed Vege-Roofing tiles

No.	Dry Weight of Tile, $W_d$	Saturated Weight of Tile, $W_s$	Absorption % (Cold Water)	Saturated weight of tile after 5 hr in boiling water, $W_b$	Absorption % (Boiling Water)	Saturation Coefficient
1	1970.5	2072.5	5.2	2110.2	7.1	0.73
2	1982.2	2076.5	4.7	2145.6	7.7	0.61
3	1968.3	2070.0	5.2	2128.1	8.1	0.64
4	2012.5	2107.4	4.7	2153.4	7.0	0.67
5	2002.6	2110.5	5.4	2141.5	6.9	0.78

462  
 463  
 464  
 465

466 **Table 10:** Embodied energy requirements in one catalyzed Vege-Roofing tile

Material	Embodied Energy (MJ/kg)	Material Required per tile (kg, L)	Total Embodied Energy per tile (MJ/kg)
Sulfuric Acid	5.00 <sup>[a]</sup>	0.006	0.03
Waste Vegetable oil	2.00 <sup>[b]</sup>	0.157	0.31
Processing	0.06 <sup>[c]</sup>	-	0.06
Sand	0.20 <sup>[d]</sup>	1.128	0.23
Fly Ash	0.00 <sup>[e]</sup>	0.608	0
			<b>0.64</b>

467 \*a, (Eric et al, 2002); b, (Reijnders and Huijbregts, 2008); c, (Francois, 2001); d, (Ecoinvent 3.3,  
468 2016); e, (Chani et al, 2003; Ostwal and Chitawadagi, 2014).

469 It is discovered from Table 10 that the embodied energy per catalyzed Vege-Roofing tile found to  
470 be 0.64 MJ/kg. This indicates that the embodied energy requirement in producing single catalyzed  
471 Vege-Roofing tiles was quite low in comparison to the embodied energy required to produce  
472 conventional clay and concrete roofing tiles (Hammond and Jones, 2008). Comparative analysis  
473 of embodied energy for similar dimensions of conventional roofing tiles and catalyzed Vege-  
474 Roofing tile is presented in Table 11. It is discovered from Table 11 that the embodied energy of  
475 catalyzed Vege-Roofing tile is 321% less than conventional concrete tile, 837% less than clay tile  
476 and 1775% less than ceramic tile as calculated by Hammond and Jones (2008). Embodied carbon  
477 in producing one catalyzed Vege-Roofing tile for different phases is demonstrated in Table 12.

478

479 Table 11: Comparative analysis of embodied energy



S. No	Tile (390mmx240mmx10mm)	Total Embodied Energy (MJ)
1	Concrete	2.7
2	Clay	6.0
3	Ceramic	12.0
4	Catalyzed Vege	0.64

480

481 **Table 12:** \*Total carbon emissions in different phases of catalyzed Vege-Roofing tile

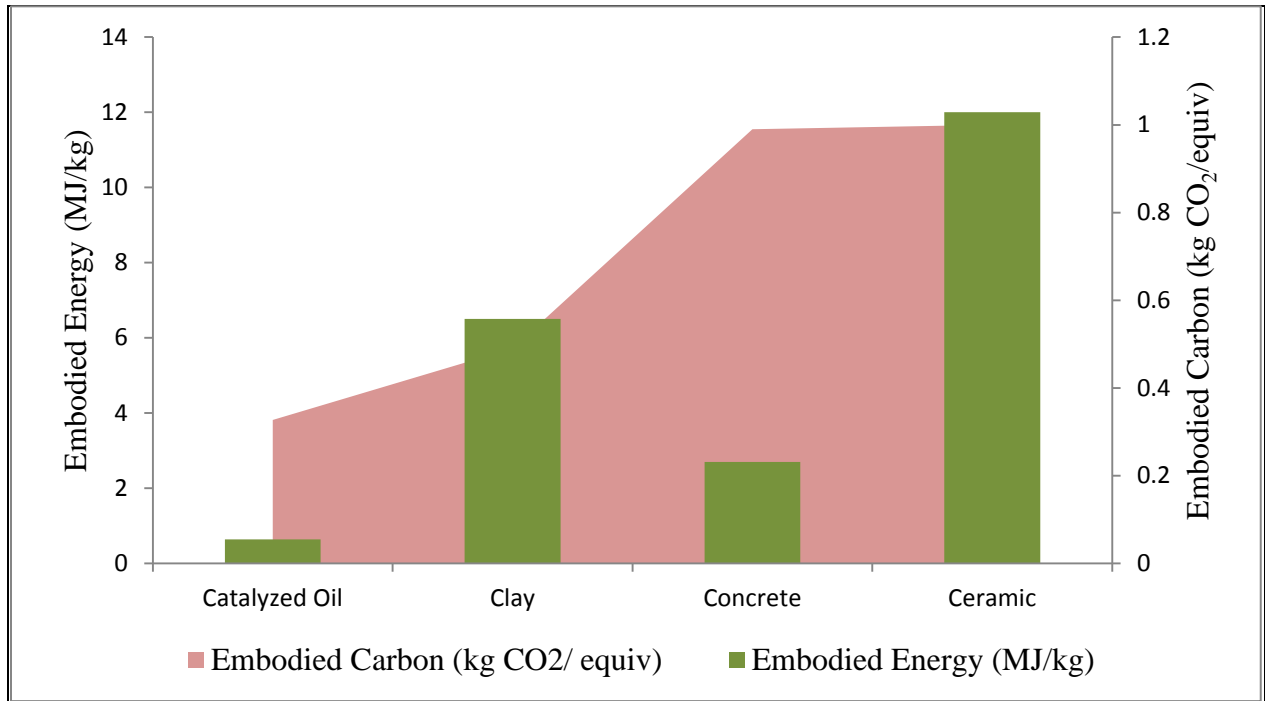
<b>Cradle to Gate Emissions</b>					
Material	Quantity (kg)	Emission factor kg CO <sub>2</sub> /equiv.	Transport (Km)	Transport	
				Emissions (per kg, Km)	Total Emission
Waste Oil	0.1574	0	1000	0.0001	0.100
Sulfuric Acid	0.0066	0.17	40	0.0001	0.005
Fly Ash	0.6080	0.004	40	0.0001	0.006
Sand	1.1280	0	20	0.0001	0.002
<b>Total Phase Emission</b>					<b>0.113</b>
<b>Manufacturing</b>					
Operation	Curing Time (hours)	Electricity Usage (KWh)	Emission Factor (kg CO <sub>2</sub> per equiv.)		<b>Total Phase Emission</b>
Heat Curing	18	0.3	0.63		<b>0.19</b>
<b>Distribution</b>					

Material	Transport (Km)	Transport Emissions (per kg, Km)	Total Phase Emission
Roofing Tile	200	0.0001	<b>0.020</b>
<b>End of Life</b>			
Material	Transport (Km)	Transport Emissions (per kg, Km)	Total Phase Emission
Roofing Tile	40	0.0001	<b>0.004</b>
<b>Total</b>			<b>0.327</b>

482 \*Emission factors values were demonstrated from Ecoinvent 3.3.

483 Table 12 reveals that estimated CO<sub>2</sub> emission value per tile for the same batch of tiles was  
484 calculated to be 0.327 kg CO<sub>2</sub>/equivalent, which is low in contrast to the traditional roofing tiles.  
485 The carbon emissions for catalyzed Vege-Roofing tiles were 202%, 49% and 205% lower in  
486 contrast to concrete, clay and ceramic roofing tiles as determined by ecoinvent 3.3. Moreover, the  
487 emission values determined for novel tiles were 267%, 37% and 126% lower than concrete, clay  
488 and ceramic tiles as estimated by Hammond and Jones, (2008). This indicates that catalyzed Vege-  
489 Roofing tiles if implemented would be environmentally friendly and will reduce the energy  
490 emissions to an appreciable amount. Comparison of energy emission values for catalyzed Vege-  
491 Roofing tiles and conventional roofing tiles is displayed in Fig. 11.

492



493

494

495 **Fig. 11: Energy Comparison of catalyzed and traditional roofing tiles**

496 Fig. 11 indicates that catalyzed Vege-Roofing tiles are more environmentally friendly since the  
 497 energy emissions discovered to be on the lower side in contrast to traditional roofing tiles. Low  
 498 energy emissions are due to the replacement of masonry units such as kiln firing in clay and cement  
 499 production (Hammond and Jones, 2008). Traditional high energy consuming binders were  
 500 replaced by waste binder i.e. waste vegetable oil that reduced the energy emissions to a remarkable  
 501 extent.

502 *3.3.4 Economic Evaluation*

503 The economy of the product is also evaluated as one of the important criterions to determine the  
 504 feasibility of the product. Cost of power and water consumption for industrial sector found to be  
 505 around RM 0.38 per KWh and RM 2.07 per cubic meter as retrieved from Tenaga Nasional Berhad

506 and Syarikat Bekalan Air Selangor SDN. BHD, Malaysia respectively. Fly ash, sand, cement,  
507 sulfuric acid and waste palm oil could be purchased from local suppliers in Malaysia at a price of  
508 RM 80, RM 80, RM 400, RM 500 and RM 400 per metric ton respectively. For cemented tile,  
509 cost was determined by using the cement to sand ratio of 1:4 (Johansson, 1995) mixed with an  
510 appropriate quantity of water. However, additional amount of water needed for the hardening  
511 process of these cemented tiles was also estimated for cost calculation. Cost comparison of same  
512 batch of catalyzed Vege-Roofing tiles and concrete roofing tiles is displayed in Table 13. Rate per  
513 tile was calculated by multiplying the material used in producing one tile with the rate of that  
514 particular material. Table 13 shows that the cost of catalyzed Vege-Roofing tile was comparatively  
515 less in contrast to conventional concrete roofing tile. Utilization of catalyst induced a major  
516 contribution to reduce the cost of the process since it reduced the curing time of the production.

517

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526 **Table 13:** Economic comparison of cemented and catalyzed Vege-Roofing tiles (a) Raw  
 527 material/tile (b) Utilities/tile

528

5 **a)**

**Cemented Tiles**

**Catalyzed Vege-Roofing tiles**

S.No.	Description	Rate (RM)	Rate per tile (RM)	S.No.	Description	Rate (RM)	Rate per Tile (RM)
1	Cement	400/metric ton	0.170	1	Fly ash	80/metric ton	0.047
2	Sand	80/metric ton	0.102	2	Sand	80/metric ton	0.077
				3	Waste Oil	400/metric ton	0.034
				4	Sulfuric Acid	500/metric ton	0.003

**b)**

1	Power Cost	0.38/KWh	0.06	1	Oven	0.38/KWh	0.12
2	Water	2.07/cubic meter	Approx 0.05	2	Mixer	0.38/KWh	0.001
		<b>TOTAL</b>	0.382			<b>TOTAL</b>	0.282

530 **4. Conclusions**

531 This research attributes to the production of catalyzed Vege Roofing tiles and discovered that  
532 catalyzed waste vegetable oil can be used as an alternate binder for the manufacturing of roofing  
533 tiles. The fabricated catalyzed Vege Roofing tiles has met the criteria for standard concrete or kiln  
534 burned clay tile as it indicated high flexural strength, low water absorption and impermeability.  
535 Implementation of novel roofing tiles would be economical and energy efficient process since  
536 energy requirements and cost are comparatively low in contrast to traditional roofing tiles.  
537 Production of this novel bio-composite also paved a way for the production of other building  
538 materials such as building blocks, flooring etc. by industrial symbiosis and thus converting the  
539 building production to a more cleaner and greener process. Additionally, this research has a scope  
540 to eliminate waste disposal problems, since fly ash, a waste from thermal power plant and waste  
541 vegetable oil, a waste from local restaurants being utilized in producing these novel tiles. **However,**  
542 **due to the formation of solid polyester as a binder in this work, UV degradation should be**  
543 **considered as a special care to use the catalyzed Vege-tile for roofing.** This novel process for the  
544 production of building materials would help in conserving the existing resources. Environmentally  
545 friendly production of building materials, low cost production and the waste management are the  
546 remarkable outcomes of this research.

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**Nomenclature**

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ASTM	American Society for Testing and Materials
BHD	Berhad (Private)
MPa	mega-pascal (unit of strength)
MJ/kg	Mega-joule per kilogram (unit of energy)

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KWh	Kilowatt hour (unit of power)
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RM	Malaysian Ringgit (currency)
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WSDOT	Washington State Department of Transportation
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