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An Attempt to Maximize the Use of Used Cooking Oil and Fly Ash in the Production of Green Roofing Tile

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Abstract: This study introduces a novel attempt of utilizing used cooking oil (UCO) and fly ash in the production of green roofing tile, namely UCO-GRT. UCO was utilized as an alternative binder to fully replace the cement and clay, while fly ash was used as a fine aggregate instead of virgin sand. This can maximize the percentage of waste substitution in the manufacturing process, consequently reducing the waste disposal issues encountered in Malaysia. The optimization process was carried out to investigate the optimal manufacturing parameters, by considering the curing duration, the composition of UCO and fly ash, and the composition of catalyst incorporated. The mechanical properties of UCO-GRT produced, including density, dry and wet transverse strength, percentage of water absorption, and permeability have been investigated. Findings reveal that the utilization of catalysed UCO and fly ash solely in the production of roofing tiles is feasible. The optimized UCO-GRT fulfilled the basic requirements of a high-profile roofing tile as per ASTM standards. The embodied carbon and embodied energy of the novel roofing tile was also studied and compared with the cementitious and clay roofing tiles.

Keywords: Used cooking oil, fly ash, roofing tile, green production

1. Introduction

Driven by the rapid urbanization and growing population in recent decades, the rate of municipal solid waste (MSW) generation increased massively without the signs of slowing down. Looking forward, the global MSW generation is expected to grow to 3.4 billion tonnes over the next 30 years [1]. Environmental impacts caused by improper waste management are increasingly becoming a major issue worldwide. A proper MSW strategy is critical for the development of sustainable and healthy communities. However, it is often overlooked, yet approximately 1/3 of waste is recycled and composted in high-income countries, while only 4% of waste is recycled in low-income countries [2]. Lack of a holistic management system and weak environmental awareness are the major factors that deteriorated the phenomenon.

In Malaysia, the most commonly used approaches for the disposal of MSW include incineration and landfilling [3]. The incineration process is an effective way to reduce the mass of the waste materials to 20 – 30% of their original volume. However, this approach is always controversial, as it would release a huge amount of gaseous pollutants, and

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consume a large quantity of energy during the treatment process [4]. Landfilling is the oldest and cheapest method of waste management, in which the MSW is simply buried in a specific location. Unlike incineration or other resource recovery systems, landfilling does not require large investments in infrastructure or personnel in system maintenance [5]. Hence, it is widespread in low-income countries, especially those with a large open scale. However, open dumps may cause several environmental issues if the landfill is set up without a proper gas recovery system. The gaseous released from the decomposition of MSW, including 50 – 60 % of methane (CH4) and 30 – 40 % of carbon dioxides (CO2) brought significant impacts to the atmosphere [6]. Leachate is another adverse environmental issue to be concerned about. The impacts of landfill leachate to the environment has forced the authorities to impose ever stricter requirements on pollution control [7].

In recent decades, the 3R waste management strategy, which is reuse, reduction, and recovery of waste materials has been widely promoted to achieve economic savings and environmental sustainability. Used cooking oil and fly ash are among the most common waste materials generated from routine activities in Malaysia. Every year, it was reported that 50,000 tonnes of UCO is generated and disposed of without proper treatment [8]. While more than 6.8 million tonnes of fly ash is generated from six electricity power stations based in Malaysia, solely for electric generation purposes [9]. To achieve sustainable development, an increasing number of researchers are looking for alternative ways to convert waste materials into value-added products. For instance, UCO can be converted into biodiesel through mineral acid pre-treatment, followed by base-catalysed transesterification process [10]. The biodiesel blends produced from UCO showed comparable functions with diesel fuel, with lower thermal efficiency and higher fuel consumption [11]. It can also be used as an asphalt rejuvenator to increase the ductility and penetration of the asphalt binder, whilst decreasing its viscosity and softening point [12]. In addition, fly ash is a famous waste that has always been recycled and reused in various applications, especially as a substitute in the production of eco-building materials. By adding 3% of fly ash in the fired brick, it was discovered that the linear shrinkage and water absorption of the product have increased by 2.5 and 3.0% [13]. Fly ash has also been used in the production of thermal insulating foam concrete [14]. The foam concrete incorporated with 50% of fly ash exhibited a comparable compressive strength to the control sample [14]. Apart from the building materials, fly ash can be used as the ingredient for mesoporous silica synthesis and zeolite synthesis [15,16].

In the previous studies, the feasibility of utilizing waste vegetable oils, fine aggregate, fly ash, and several additives in the production of roofing tile were carried out [17,18]. The usage of cement and clay that possessed high embodied carbon and embodied energy has been terminated throughout the study [19]. Generally, the eco-roofing tiles produced from those studies had convincing performance which can fulfil the requirements of a high-profile roofing tile in accordance with ASTM standards [20]. However, in the previous study, the replacement of conventional ingredients by 35 – 40 % of waste materials in the production of innovative green roofing tiles is still unsatisfactory. The product still needed high composition of fine sand to enhance its mechanical properties. To maximize the usage of waste material product, a bold idea of utilizing only waste materials in the production of roofing tiles was attempted. The production of a fully waste-made building material provides an alternative approach for the management of waste, reduces the consumption of natural resources, and enhances the industrial value of the waste products.

2. Materials and Methodology

2.1 Raw Materials

2.1.1 Used Cooking Oil (UCO)

Used cooking oil utilized in the production of UCO-GRT is obtained from the local restaurants. UCO is a brownish liquid with a viscosity of 168.8 cP and specific gravity of 0.92. Theoretically, UCO possessed a significant amount of glycerol, vast ranges of free fatty acids, secondary oxidation products, and various impurities. However, the composition of UCO might vary according to the degradation extent of UCO during the cooking and frying process. Hence, the collected UCO from the different restaurants were mixed thoroughly to enhance the consistency of the result obtained. **Figure 1** shows the physical appearance of virgin and used cooking oil.

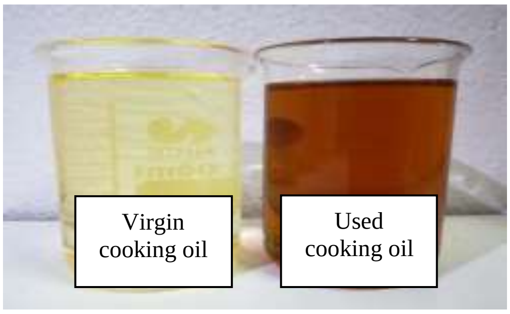

Fig. 1 - Physical appearance of virgin and used cooking oil

2.1.2 Fly Ash

Fly ash is a brownish waste material generated from the thermal power plants, TNB Janamanjung Sdn. Bhd. located in Perak, Malaysia. It is served as the major ingredient of the UCO-GRT. The collected fly ash was oven-dried at 105 ^{o}C and sieved to eliminate the unburnt components to ensure homogeneity. The average particle size of the fly ash in terms of number and volume was analyzed using the Particle size analyzer, *Mastersizer 2000*, which is determined to be 2.124 µm and 14.415 µm respectively. The specific gravity of fly ash is 0.26.

2.1.3 Sulfuric Acid

Sulfuric acid, H_2SO_4 was used as a catalyst to enhance the rate of polymerization reactions of UCO upon the thermal treatment. In this study, the AR grade of concentrated sulfuric acid with an original concentration of 8.33M was diluted to 0.1M through a simple dilution method before the mixing process with the UCO.

2.2 Manufacturing Process of UCO-GRT

In this study, only used cooking oil (UCO) and fly ash were utilized in the production of a novel roofing tile, namely UCO-GRT. UCO was utilized as the alternative binder, while fly ash was incorporated as the natural aggregate in the manufacturing process. This approach avoided the usage of cement and clay, which are considered non-eco-friendly ingredients as they possessed high embodied carbon and embodied energy. In addition, the addition of sand aggregates is also avoided to maximize the extent of waste replacement in the production of UCO-GRT.

Figure 2 shows the manufacturing process of the UCO-GRT. The manufacturing process consists of three steps, which are mixing, compacting, and heat curing process. In the mixing process, a specific composition of UCO and fly ash were mixed thoroughly in the bench mounting mixer for 5 – 10 minutes to ensure all the ash particles are well encapsulated by the UCO. After being thoroughly mixed, the resultant mixture was poured into the mold and compacted using a Marshall compactor. The UCO-GRT will be produced in two different sizes. For optimization purposes, the samples will be produced in a round shape with 100 mm in diameter and 25 mm in thickness. While for the verification process, the UCO-GRT was fabricated in the standard size of 390 × 240 × 25 mm. Each sample was compacted 20 times. Lastly, the compacted mixture was off-molded, and heat cured in a ventilated oven at 190 ^{o}C. After 12 – 30 hours of curing durations, the rigidified samples were taken out and cooled down at the ambient temperature. All the samples produced were then proceeded to a series of mechanical tests, including the transverse strength, percentage of water absorption, and permeability tests.

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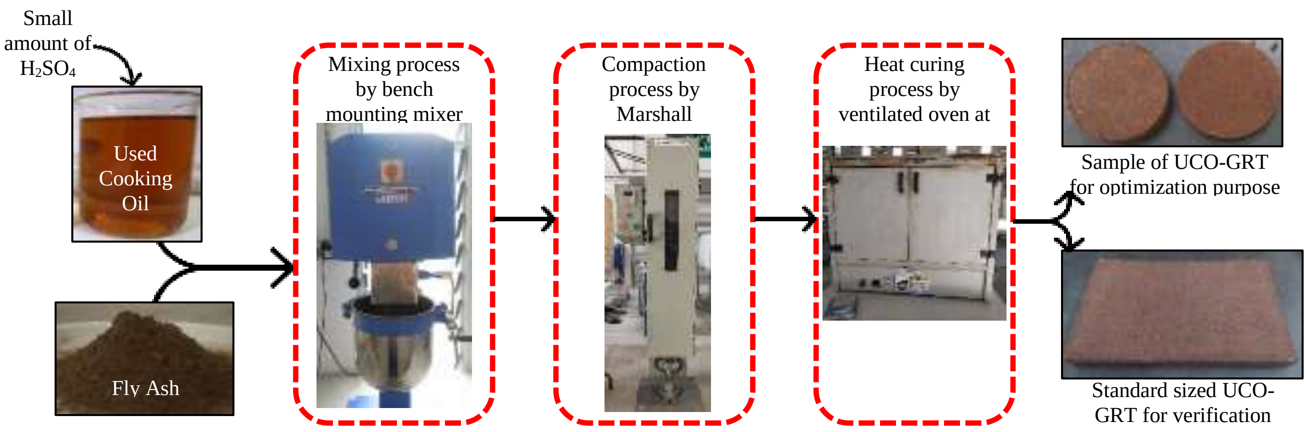

Fig. 2 - Manufacturing process of UCO-GRT

2.3 Testing Procedures

2.3.1 Dry and Wet Transverse Strength

The transverse strength of the samples was determined according to the procedures outlined in ASTM C 1167 – 03 [21]. It can be categorized into dry and wet transverse strengths. For the dry transverse strength test, the samples need to be oven-dried at 105 °C prior to the analysis. For the wet transverse strength test, the samples were immersed in distilled water for 24 hours prior to the test. All the tests were carried out in the three-point bending mode by using a Materials Testing Machine (T-machine, model LTSH-50KN) equipped with U.T.M Operation Program for data evaluation.

2.3.2 Percentage of Water Absorption Test

The water absorbability of the sample was determined in accordance with the ASTM C 67 – 07a [22]. The dry and wet mass of the samples were measured after oven-dried or immersed in water for 24 hours. The percentage of water absorption and saturation coefficient for a specimen can be calculated as follows:

$$Percentage\ of\ Water\ Absorption, \% = [(W_w - W_d)/W_d] \times 100\%$$

where: W_d = dry mass of sample after oven-dried at 105 °C for 24 hours

W_w = wet mass of sample after immersed in water for 24 hours

3.2.3 Permeability Test

The permeability test is carried out in accordance with ASTM C 1492-03 [23]. An open-bottom trough that covered 80% of the surface of the specimen was fixed horizontally on the samples. Distilled water was poured into the trough for at least 50 mm height above the sample. The sample was left on a supporting frame. The underside of the sample was observed periodically for signs of water droplets. No penetration of water on the underside of the specimen after 24 hours duration is indicative of impermeability of the tiles.

3.0 Results and Discussion

3.1 Manufacturing Process of UCO-made roofing tiles

This section discusses the optimization process of the manufacturing parameters of the UCO-GRT. All the samples of UCO-GRT were tested for their dry and wet transverse strengths, percentage of water absorption, and permeation characteristics. The sample which possesses the greatest mechanical strength, whilst fulfilling the requirements of water absorbability and impermeability, was justified as the optimized UCO-GRT. The manufacturing parameters were being optimized at the end of this section. The manufacturing optimized parameters included the curing duration, binder and filler composition, and the composition of catalyst incorporated. The initial appropriate parameters for the manufacturing of the samples are 5% of UCO, 95 % of fly ash, 190 °C of curing temperature, and 24 hours of curing duration. The variety of each parameter was adjusted in accordance with the above-mentioned parameters.

3.1.1 Optimization of Curing Duration

At the beginning of the optimization process, the composition of UCO, composition of fly ash, and curing temperature used to produce the sample of UCO-GRT were fixed at 5%, 95%, and 195 ^{o}C respectively. Total curing duration of the UCO-GRT was between 12 to 30 hours. **Figure 3** shows the mechanical strength of samples produced from different curing durations. From the result obtained, the strength of samples produced between 16 to 24 hours of curing duration fulfilled the requirement of dry transverse strength of 1779 N as outlined in ASTM C 1167 – 03 [21]. The greatest strength achieved by UCO-GRT happened when it was produced under 18 hours of curing duration. However, the prolonged curing duration led to the descending of the mechanical strength of the samples. The is attributed to the presence of excessive heat energy in the samples, which may cause the deterioration of the chemical structure of the polymerized UCO components [24]. The deterioration process brought significant impacts on the binding matrix, consequently weakening the strength of the samples. In addition, longer curing duration also would increase the cost of the manufacturing process, as well as the embodied carbon and embodied energy of the samples. Hence, curing duration of 18 hours is considered as the optimized curing duration for the manufacturing of UCO-GRT.

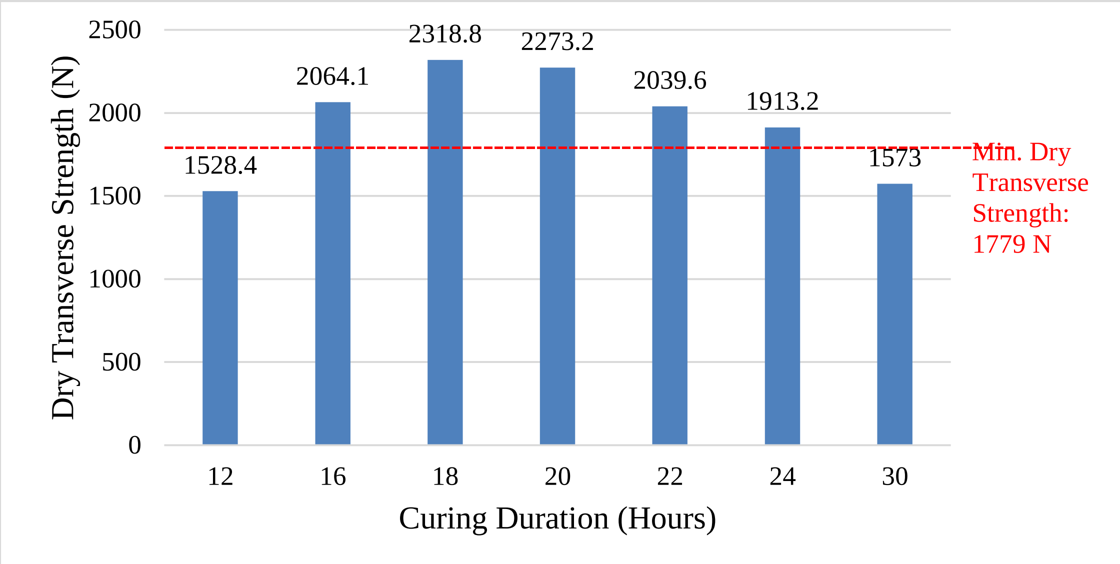

Fig. 3 - Dry transverse strength of UCO-GRT produced from different durations

3.1.2 Optimization of UCO Composition

In the rigidification process of UCO-GRT under the elevated temperature, the chemical compositions of UCO, including glycerol, free fatty acid, and secondary oxidation products took part in the complex oxy-polymerization reaction and further contributed to the strength development process [17]. It was noticed that the composition of UCO plays an important role in the manufacturing process as it corresponds to the mechanical strength of the samples. As an effort to maximize the strength achieved, a series of samples were produced by using different proportions of UCO to fly ash, ranging from 4.0 – 8.0 %, with 0.5 % intervals. While the curing temperature and duration were fixed at 190 ^{o}C and 18 hours respectively. The average strength achieved by the samples was determined and shown in **Figure 4**. From the data obtained, most of the samples achieved the strength requirements as per ASTM C 67-03a, with an exception when only 4% of UCO is utilized. The sample produced from 4% of UCO possessed the lowest transverse strength, which is 1594.5 N. Along with the addition of the UCO composition, an increasing trend in the strength of samples was indicated. This indicated that UCO-GRT with higher UCO concentration would have better binding matrix and mechanical strength. The greatest strength of 3447.0 N was achieved by UCO-GRT with 6.5% of UCO concentration.

However, the dry transverse strength of UCO-GRT decreased when its UCO concentration was beyond 7.0%. This may be due to more heat energy being required to trigger a complete oxy-polymerization reaction of the higher concentration of UCO-GRT. Within the limited curing duration of 18 hours, the incomplete chemical reactions consequently resulted in a lower mechanical strength. When a greater amount of UCO is added, the mechanical strength of the specimen would be lower. Hence, UCO composition of 6.5% was determined as the optimized value in the manufacturing of UCO-GRT.

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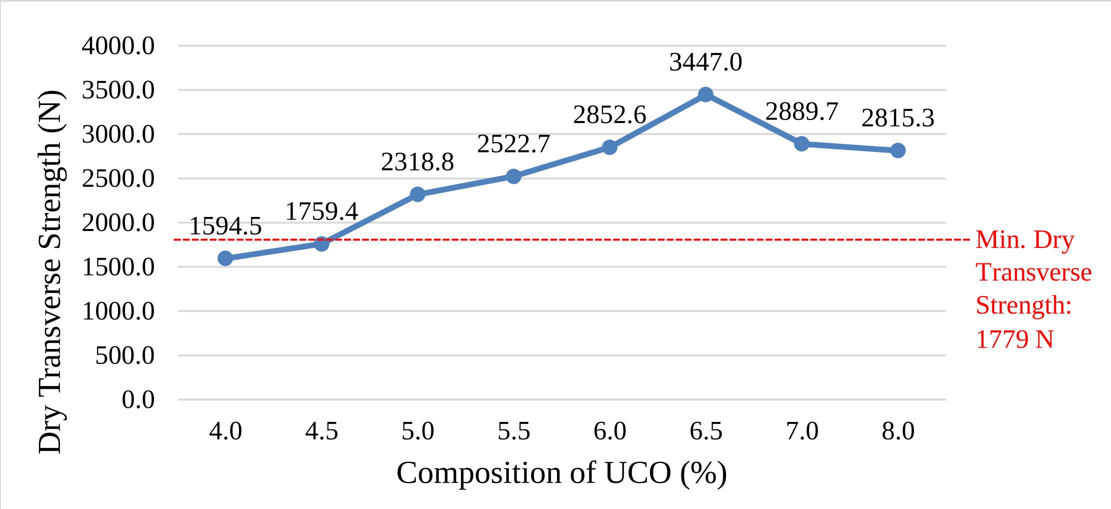

Fig. 4 - Average dry transverse strength and flexural strength achieved by UCO-GRT produced with different composition of binder and filler content

3.1.3 Optimization of the Catalyst Composition

In the previous study, it was revealed that sulphuric acid, H_2SO_4 possessed the greatest performance when utilized as an enhancer in promoting the oxy-polymerization reactions of waste oil [24]. This is because sulfuric acid tends to release two hydrogen ions (H^+) to promote polymerization reactions. In the presence of sufficient catalysts, the rate of reactions can be enhanced to a significant extent. Hence, H_2SO_4 was chosen as the catalyst throughout the optimization process. In this section, a specific amount of H_2SO_4 was added into and mixed thoroughly with UCO prior to the mixing process with the fly ash. The composition of the catalyst incorporated is ranging from 0.25 to 2.00 % with respect to the total weight of the sample. The other manufacturing parameters were fixed at 6.5% of UCO, 93.5% of fly ash, 190 ^{o}C of curing temperature, and 18 hours of curing duration.

Figure 5 shows the mechanical strength achieved by a series of samples incorporated with different compositions of catalyst. Obviously, all the samples produced possessed mechanical strength that far exceeded the minimum requirement in accordance with ASTM standards. In addition, the mechanical strength of the samples was also enhanced significantly with the addition of H_2SO_4. When 1.25 % of H_2SO_4 was incorporated into the sample, the greatest strength of 4256.7 N was achieved. When a greater amount of UCO was introduced, it was expected that more catalysts were needed to trigger and accelerate the oxy-polymerization reaction. However, the addition of a catalyst exceeding the optimal value leads to a decrease in the samples' strength. This is because the catalyst would not be exhausted at the end of the chemical reactions, whilst not contributing to the strength development of samples. Hence, the presence of excessive catalyst obstructed the bindings between the UCO and fly ash particles, subsequently decreasing the strength of the roofing tiles.

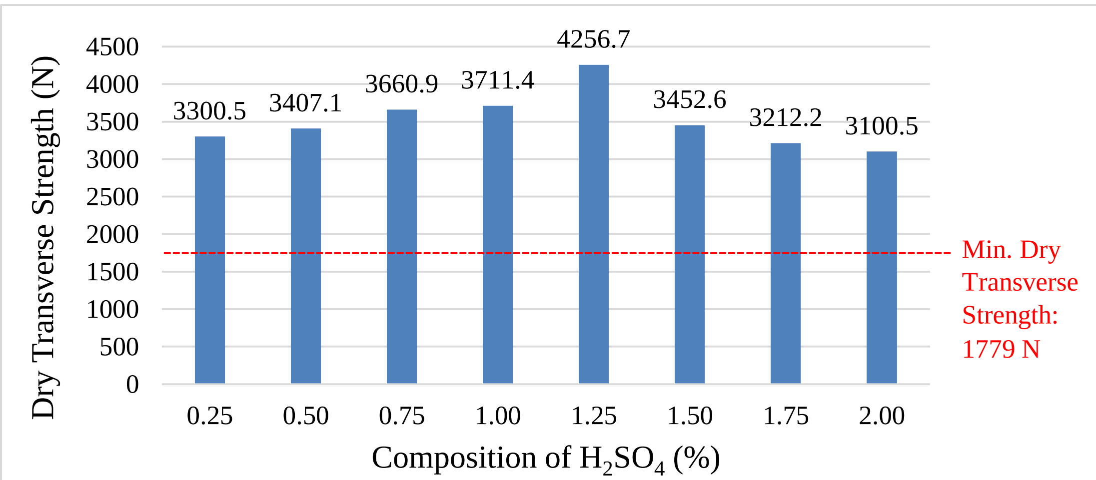

Fig. 5 - The dry transverse strength of UCO-GRT incorporated with different composition of H_2SO_4

3.2 Verification Process of UCO-GRT

Based on the results obtained from the optimization processes, the optimal manufacturing parameters which were expected able to produce a UCO-GRT with the greatest properties are as follows:

(a) 6.5% of UCO and 93.5% of fly ash
(b) 18 hours of curing duration
(c) 0.0125% of H_2SO_4 as the catalyst

In this verification process, triplicate UCO-GRT were prepared in the standard size with the dimension of 390 mm × 240 mm × 25 mm using the above-mentioned parameters. The density, dry and wet transverse strengths, percentage of water absorption, and permeable characteristics of the optimized UCO-GRT were determined. This verification process would provide a better view of the potential of UCO and fly ash in the production of fully waste-made roofing tile.

According to the American Concrete Institute, ACI 213R, the average density of lightweight and normal-weight cementitious building materials is between 1.440 – 1.840 g/cm^3 and 2.240 – 2.400 g/cm^3 respectively. **Table 1** shows the average density of optimized UCO-GRT, which is 1982 ± 12 kg/m^3. Hence, this suggests that UCO-GRT produced in this study can be categorized as a normal-weight building material. In addition, the mechanical properties of the UCO-GRT are also shown in Table 1. Overall, the optimized UCO-GRT fulfilled all the basic requirements of a roofing tile in accordance with ASTM standards. The average dry transverse strength achieved by the UCO-GRT is 4314.7 ± 101.6 N, which is comparable with the result obtained through the optimization processes. The optimized UCO-GRT can also be categorized as a high-profile roofing tile as its mechanical strength is far above the requirements of 1779 N.

The average percentage of water absorbed by the optimized UCO-GRT is 5.24%, fulfilling the limitation of 6% as per the ASTM standards. This is attributed to the hydrophobicity of UCO, which exhibited water-repelling effects to reduce water penetrating rate of the UCO-GRT. Nevertheless, a certain amount of water molecules is still able to penetrate and adhere to the fly ash. This condition can be minimized when all the ash particles are well-encapsulated by the UCO through the mixing process. The encapsulation of ash particles by UCO components prevents them from being directly exposed to the water molecules, consequently reducing the water absorbability of the UCO-GRT [20]. The low percentage of water absorption of UCO-GRT resulted in a better wet transverse strength of UCO-GRT, which is 3288.1 N, fulfilling the standard as a high-profile roofing tile. Lastly, UCO-GRT is impermeable. It is well compacted by the Marshall compactor, hence successfully restricting the penetration of water molecules through the sample over 24 hours of experimental duration.

Table 1 - Mechanical properties of the optimized UCO-GRT

Mechanical properties	Achieved value	Requirements (ASTM standards)
Density	1982 ± 12 kg/m^3	1.440 – 1.840 g/cm^3 = lightweight 2.240 – 2.400 g/cm^3 = normal weight
Dry transverse strength	4314.7 ± 101.6 N	≥ 1779 N
Percentage of water absorption	5.24 ± 0.68 %	≤ 6.0 %
Wet transverse strength	3288.1 ± 107.1 N	≥ 1334 N
Permeability	Pass	No penetration of water after 24 hours

3.3 Life Cycle Assessment of the Optimized UCO-GRT

The life cycle assessment of UCO-GRT in terms of embodied carbon and embodied energy (EC & EE) was carried out and compared with the conventional products. **Figure 6** shows the comparative study of the EC & EE of UCO-GRT, concrete, and clay roofing tiles. Generally, the embodied carbon of the UCO-GRT was analysed through its carbon emissions during the materials extraction and products fabrication (cradle to gate), products distribution (cradle to site), and end-of-life management (cradle to grave) [25]. While the embodied energy of UCO-GRT was estimated through the energy consumed throughout the manufacturing process. As a result, the EC & EE of UCO-GRT were determined as 105 kgCO_2/tonnes and 156 MJ/tonnes respectively, which are much lower compared to the conventional roofing tiles. This is because the waste materials, such as UCO and fly ash have lost most of their invented carbon during their service life [26]. Hence, UCO and fly ash possessed lower carbon emission factors, ranging between 0.000 – 0.004 kgCO_2/kg [17]. Full replacement of conventional raw materials, such as cement and clay which consists of significant carbon footprints tends to significantly reduce the embodied carbon of the UCO-GRT. It was discovered that the embodied carbon of UCO-GRT is 53.1 % and 77.2 % lesser than cementitious and clay roofing tiles respectively. In addition, the embodied energy of the UCO-GRT is 90.5% and 97.6 % lower than concrete and clay roofing tiles too. This is because the production of cement clinkers and the clay-firing process require a temperature up to 1000 ^oC which consumes a large quantity of energy during the manufacturing process. The replacement of these traditional materials,

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as well as the curing temperature of 190 °C in the production of UCO-GRT, is considered reasonable and energy-saving. Hence, the energy consumed throughout the manufacturing process of UCO-GRT was reduced to a significant extent. Conclusively, the utilization of UCO and fly ash in the production of fully waste-made roofing tiles could be classified as an environmentally friendly building material.

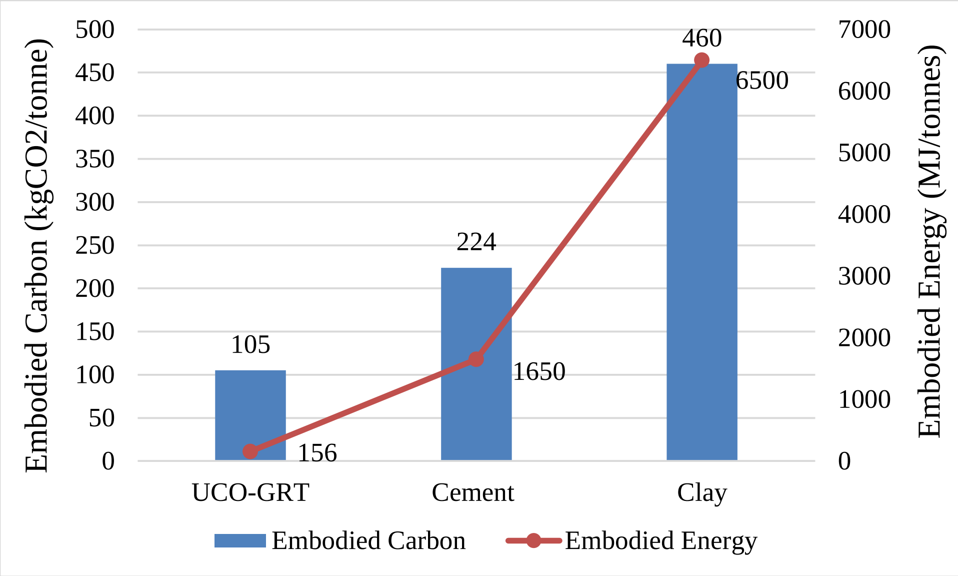

Fig. 6 - Comparison of the embodied carbon and embodied energy of UCO-GRT, concrete and clay roofing tiles.

4.0 Conclusion

In this study, utilizing only UCO and fly ash in the production of roofing tiles is proven feasible. Through a series of optimization processes, it was found that 6.5% of UCO and 93.5% of fly ash, 18 hours of curing duration at 190 °C and adding 1.25 % of H_2SO_4 as a catalyst can produce the UCO-GRT with optimal properties. The maximum dry and wet transverse strength achieved by the optimal UCO-GRT are 4314.7 N and 3288.1 N respectively, fulfilling the requirements in accordance with ASTM C 67-03a, C 1167-03, and C1492-03. The optimized UCO-GRT also possessed low water absorbability of 5.24 %.

In terms of environmental perspective, UCO-GRT possessed relatively lower embodied carbon and embodied energy compared to conventional building materials. This attempt successfully enhances the waste replacement extent in the production of roofing tiles up to 99%. Hence, it paved an alternative option for the reduction of waste disposal issue, reduction of the consumption of virgin materials, whilst culminating in sustainable development and cleaner production.

5.0 Acknowledgement

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