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Utilization of ethyl cellulose polymer and waste materials for roofing tile production

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Abstract. The aim of this study was to utilize ethyl cellulose, mixture of waste engine oil and waste vegetable oil as a binder in the environmental friendly roofing tile production. The waste engine-vegetable oil was mixed together with ethyl cellulose, fly ash, coarse aggregates, fine aggregates and a catalyst. The Fourier Transform Infrared (FTIR) analysis showed that the oil mixture added with ethyl cellulose has the relatively high binding effect due to the presence of strong carbonyl group especially after being heat cured at 190°C for 24 hours. The mixed proportion of materials with different amount of ethyl cellulose used was studied in the production of tile specimen. The results showed that the ethyl cellulose composed roofing tile specimens passed the transverse breaking strength, durability, permeability and the ultraviolet accelerated test. The shrinkage on the tile can be overcome by adding temperature resistance polymer on the exterior of the tile.

1. Introduction

Knowledge of the use of recycled materials in the production of roofing tiles can facilitate the development of green technologies for building construction. Ethyl cellulose is a derivative of cellulose in which some of the hydroxyl groups on the repeating glucose units were converted into ethyl ether groups as shown in Figure 1. Ethyl cellulose offers an attractive range of physical properties and blended with other materials to achieve intermediate characteristics like roofing tile that used as a binder. It provides a green strength to the unfired roofing tile and burnt out clearly leaving no residue after firing. Ethyl cellulose polymer is inert and high purity powders with no caloric value. Ethyl cellulose polymer has been widely used in applications like pharmaceutical, personal care, food, specialty applications and others due to their physical and chemical properties [1]. Ethyl cellulose is used as a binder in some clay products, gives good binding strength, and fires out of the ceramic with an ash residue of only 0.5%. In addition, clays have been used for millennia and can obtain form stability even without being fired. The unfired clays were vulnerable to water and had never been used to form food and beverage containers which have utility in the fast food industry. Nevertheless, if clay is included in large enough amounts, it can impart some degree of binding to the hydrated mixture [2]. Emulsions are obtained at 30°C whereas strong concentrated oil-in-water emulsions are obtained at below 30°C. Ethyl cellulose functions as a polymeric emulsifier. The high oil-linking tendency of these particles is due to the accelerating temperature of ethyl cellulose in the lipid [3]. A phase separation method is used to mix the rosemary oil with ethyl cellulose microcapsules. A regular spherical shape is observed on the ethyl cellulose microcapsules. The stirring speed of the encapsulation affected the size and degree of sphericity of the microcapsules [4]. The polymer ethyl



cellulose is mixed with vegetable oil at the temperature above 140°C to induce gelation. Oil oxidation and the breakdown of ethyl cellulose occurred at high temperature [5]. The vegetable oil is blended with 1 wt % of ethyl cellulose at a temperature between 100 and 150°C at 300 rpm for 5 to 10 h. The mixing is influenced by the concentration and nature of additive. The heat is required to dissolve the ethyl cellulose in the vegetable oil [6].

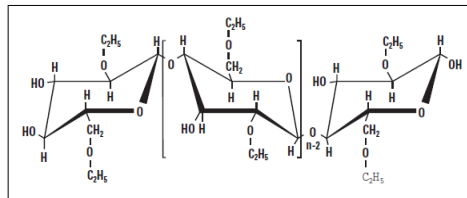


Figure 1. Chemical structure of ethyl cellulose.

Heat sonication treatment is a highly effective way for physical, chemical and enzymatic modification of different starches. Heat sonication process is conducted to promote de-agglomeration and dispersion of the mixture of starch and oil and therefore, producing a homogeneous mixture. In addition, the process modified the molecular structure of the starch without the usage of any chemicals. The oil-polysaccharide mixtures prepared earlier are sonicated using a Cole-Parmer stainless steel ultrasonic tank. The sonicator bath is filled with water until the water level reached one inch from the top of the tank. The oil-polysaccharide mixtures are sonicated with water heated to 60 °C under a frequency of 40 kHz for 20 min [7]. The objectives of this study are to optimize the tile compositions and manufacturing processes. In addition, the mechanical properties, durability and permeability of the tile also have been studied.

2. Materials and method

2.1 Preparation of the materials

Waste engine oil and waste vegetable oil were collected from local car station and restaurant respectively. The waste engine-vegetable oil was blended in the ratio 1:3 and different amount of ethyl cellulose powder were added to the mixture in an orbital shaker at 300 rpm at 100°C for 30 minutes to dissolve it. The ethyl cellulose polymers used were from ETHOCEL branded from Sigma-Aldrich. The river sand was used in the mixture to produce tiles. The size of river sand used was between 5 to 10 mm and below 5 mm which was obtained from sieving. The type of fly ash used was class F fly ash with the size of less than 75 μm. Fly ash was obtained from TNB power plant station in Manjung Setiawan, Perak, Malaysia. The chemical substances in fly ash are silica, alumina, magnetite and Fe₂O₃ (Iron oxide). The spherical fly ash has specific gravity and specific surface area between 2.5 and 200 m²/kg respectively. The percentage of each material in the composition has been chosen due to easy mixing and manufacturing of the tile. The percentage of coarse sand aggregate in the composition should be less than fine aggregate for easy formation of the tile after mixing with appropriate amount of waste oil with ethyl cellulose. The fly ash was used as the filler in the composition. The fly ash has been used in brick making factories in China with low volume between 10% and 30% [8].

2.2 Characterization of the ethyl cellulose mixed with waste engine-vegetable oil in the ratio 1:3 by FTIR analysis

The 0.006%, 0.012% and 0.023% of ethyl cellulose in the mix proportion were blended in a mixture of waste engine-vegetable oil at 300 rpm at 100 °C for 30 minutes and allowed to cool down. Fourier transform infrared spectroscopy (FTIR) of model spectrum (Shamadzu, 73093) was used to study the properties of the oil. A drop of the oil was placed on the top of the soaking plate, covered and placed in the holder and analysis performed. FTIR analysis determines the availability of the base oil and

other components by spectra. Therefore, the additives, oxidation products, contaminants and breakdown products can be known [9]. The FTIR spectroscopy is important in identifying the molecular structures and the possibility to assign certain absorption bands related to the functional group of the compounds [10].

2.3 Tile production

Different amount of ethyl cellulose had been mixed with 4% of waste engine-vegetable oil in an orbital shaker at 300 rpm at 100°C for 30 minutes to dissolve it. The amounts of ethyl cellulose used were 0.006%, 0.012% and 0.023% in the compositions. The waste engine-vegetable oil which composed of ethyl cellulose was blended with 81% of river sand and 14.5% of fly ash and with the addition of 0.17% of sulfuric acid. After mixing of the materials, the weight of the materials was measured 400g before compaction by Marshall Compacter to remove the spaces and air in the mixture. The cylindrical shape tile with 100 mm diameter and 20 mm thickness was transferred to the oven for heat curing process at 190±2°C for 24 h. The composition of different amount of ethyl cellulose in the mixture is shown in Table 1. Triplicate tile specimens were prepared with varying content of ethyl cellulose in the mix proportion and were tested for transverse strength, water absorption and water permeability [11]. A test of shrinkage was carried out according to the CNS 3299 R3071 (Methods of Test for Ceramic Tiles).

Table 1. Compositions of the materials before heat curing (% weight).

Composition	5mm-10mm aggregates	Below 5mm aggregates	Fly ash	Wasteoil	Sulphuricacid	Ethyl cellulose powder	Total
1	5.80	75.46	14.51	4.054	0.17	0.006	100
2	5.80	75.46	14.51	4.049	0.17	0.012	100
3	5.80	75.46	14.51	4.037	0.17	0.023	100

3. Results and discussion

3.1 Water adsorption

The cold water adsorption test was performed according to ASTM C 1167 standard specification for clay roof tiles. The dry tile specimens were weighted and submerged in water for 24 hours and weighted again to calculate the cold water adsorption percentage.

The water absorption was calculated by using the equation:

$$\text{Absorption, \%} = (W_w - W_d) / W_d \times 100\%$$

where, W_w = wet weight (g); W_d = dry weight(g)

Results showed that the tiles had water absorption below than 5%. The water adsorption percentage of 0.006%, 0.012% and 0.023% of ethyl cellulose in the mix proportion were 4.2, 4.3 and 4.3 respectively as shown in Figure 2. The water adsorption values increased as the ethyl cellulose usage increased. The waste oils which functions as a binder prevent the entry of water molecules in the tile specimen and thus resulted in the low water adsorption percentage.

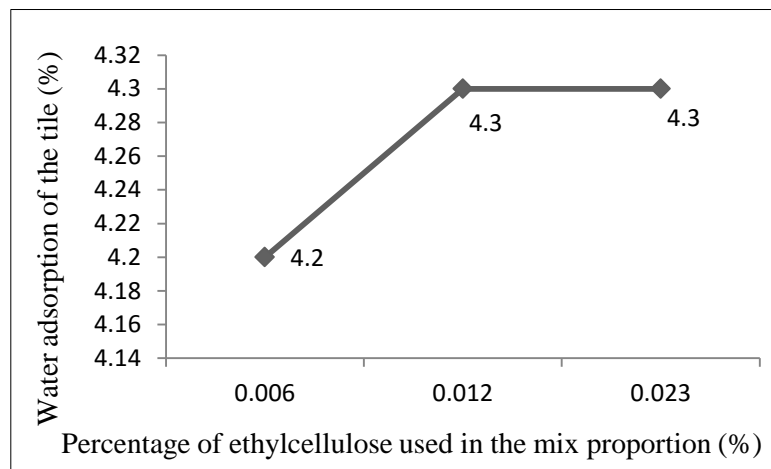


Figure 2. Cold water adsorption of the tile.

3.2 Transverse breaking strength

Transverse breaking strength test was conducted according to ASTM C 1167 standard specification for clay roof tiles. Transverse breaking strength is stated in Newton. Dry transverse breaking strength was performed by exposing the tile to heat at about 110⁰C for 24 hours before being tested for its strength using materials testing machine (Instron4469). In contrast, wet transverse breaking strength was carried out by immersing the tile into the water at room temperature for 24 hours before being tested for its strength by using materials testing machine (Instron4469). The effect of ultraviolet on the tiles was carried out by placing the tiles inside the ultraviolet chamber for 30 days. The tiles were sprayed with water five times a week to imitate the real environment of open air. After 30 days, the tiles were tested for their transverse breaking strength.

Results revealed that 0.023% of ethyl cellulose in the mix proportion showed highest dry transverse breaking strength and wet transverse breaking strength of 2510.7 N and 2234.5 N respectively as shown in Figure 3 and Figure 4. The tile specimens composed of 0.023% of ethyl cellulose in the mix proportion showed the highest transverse breaking strength of 3162 N and 2247 N for with and without the exposure of ultraviolet light respectively as shown in Figure 5. The free radicals created by the ultraviolet radiation react with the oxygen present to form hydro-peroxides that can result in polymer chain breakage [12]. From the FTIR analysis, the carbonyl that found in the ethyl cellulose that mixed with waste engine-vegetable oil in the ratio 1:3 enhance the strong binding with other materials and therefore resulted in the increase in transverse breaking strength of the tile specimen.

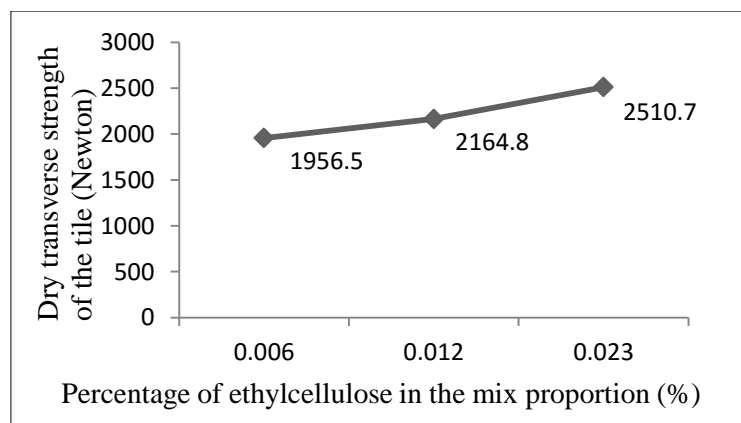


Figure 3. Dry transverse breaking strength of the tile.

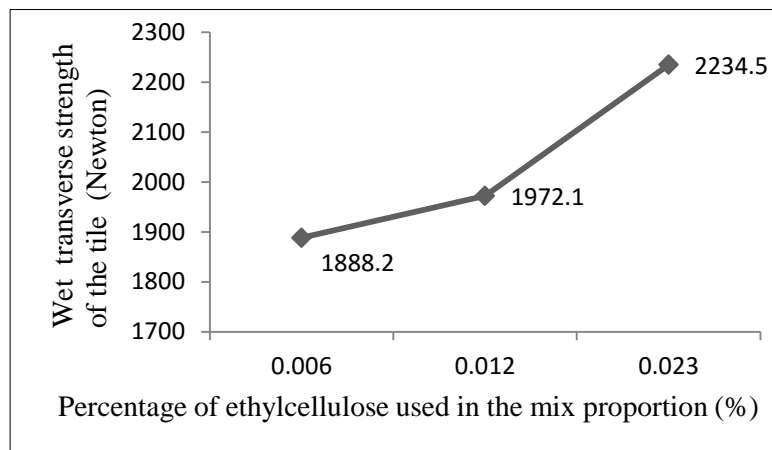


Figure 4. Wet transverse breaking strength of the tile.

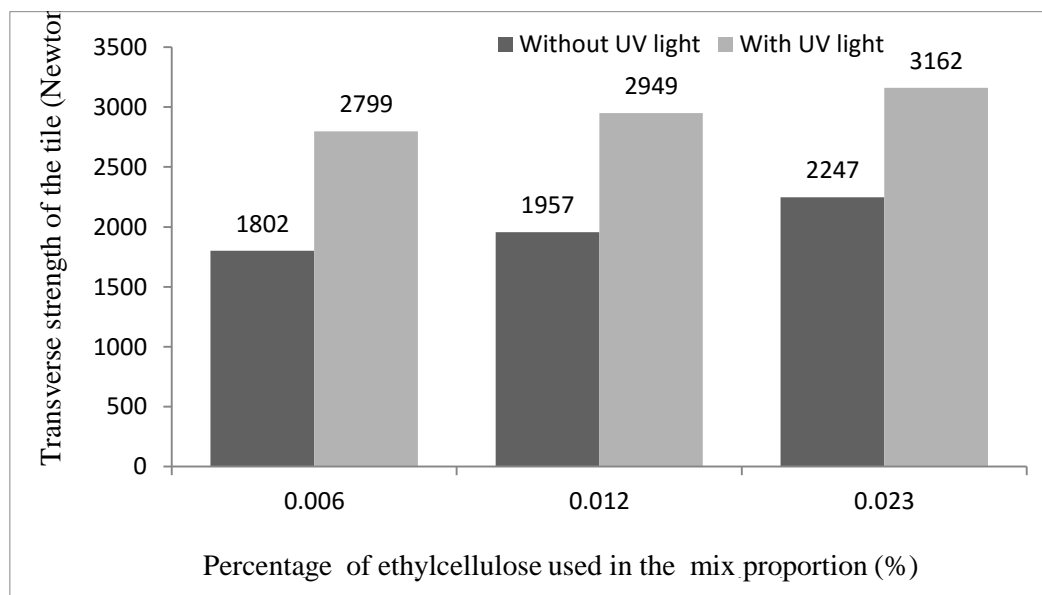


Figure 5. Transverse breaking strength of the tile (with or without ultraviolet light exposure).

3.3 Water Permeability

The water permeability test was carried out according to ASTM C 1167 Standard Specification for Clay Roof Tiles. The tile specimen was placed at the open bottom of a container filled with water at the height of 5 cm. The permeability showed the effectiveness of the concrete to prevent the movement of water into the cement [13]. Results showed that the tile specimens were impermeable to water due to the tile specimens that eliminate water from entering the tile.

3.4 Test of Shrinkage

The production of the tiles composed of ethyl cellulose and other materials requires low temperature of 190°C. However, the shrinkage test of the tiles was measured at temperatures at 1050°C, 1100°C, 1150°C and 1200°C according to CNS Regulation 3299. The shrinkage test usually was performed on ceramic tiles because ceramic tiles require high temperature between 1000°C and 1200°C during production [14]. The ceramic tiles could not produce at low temperature about 200°C. The percentages

of firing shrinkage in length of the tiles were between 30% and 40% for the tiles composed of ethyl cellulose and other materials.

3.5 Fourier Transform Infrared Spectroscopy

FTIR analysis showed that the oil mixture added ethyl cellulose has the relatively high binding effect due to the presence of strong carbonyl group especially after being heat cured at the temperature of 190°C for 24 hours. The peaks were noticed around 1746 cm^{-1} and 1747 cm^{-1} due to the presence of carbonyl such as ester, ketones and carboxylic acids as shown in Figure 6. The oxidation of oil occurred with the presence of free radicals during initiation and propagation steps in a chemical reaction [15].

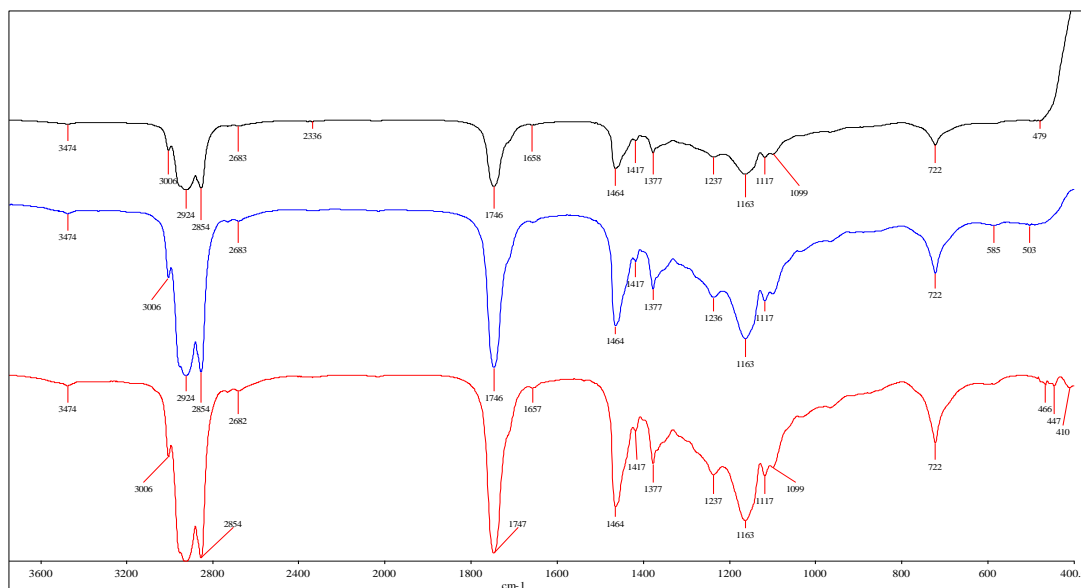


Figure 6. FTIR spectra of 0.023%, 0.012% and 0.006% concentration of ethyl cellulose in ascending order.

4. Conclusion

The tile specimens composed of 0.023% of ethyl cellulose in the mix proportion had the highest dry transverse breaking strength and wet transverse breaking strength of 2510.7 N and 2234.5 N respectively. The tile specimens had water adsorption capacity less than 5% and also impermeable to water. The results showed that the ethyl cellulose composed roofing tile specimens passed all the tests as per ASTM requirements and also passed the ultraviolet accelerated test. The ethyl cellulose roofing tile can be commercialized as an environmental friendly roofing tile with proper coating of polymer that has resistance to high temperature.

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