Study of rubber composites between natural rubber and Mahogany Shell Powder (MHSP) and potential for pavement block

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Abstract. This research was aimed to study the feasibility of using mahogany fruit shell powder as filler in rubber and investigated in properties of obtained vulcanized rubber. As mahogany fruit shell contains 75.61% holocellulose, 13.54% lignin and 11.02% extractives [1]. Hence the fruit shell should be able to act as reinforcement in rubber materials [2]. In this study, mahogany fruit shell was milled into small particles with the particle size of approximately 60 micrometres follow by ASTM D 422 - Standard Test Method for Particle-Size Analysis of Soils. The particle was characterized for their thermal stability. The morphology of mahogany fruit shell powder, MHSP, was flake like particle and can be used as reinforcement fillers in natural rubber. As NR is generally sensitive to O₂ thus ethylene propylene diene monomer, EPDM, would be laminated on the top of NR/MHSP composites. The rubbers were formulated with efficient vulcanization system and contain 10, 20, 30, 40 and 50 phr of MHSP. The rubber compounds were characterized for cure characteristic parameter such as scorch time, cure time, minimum and maximum torque. Rubber vulcanizates were prepared using compression molding and characterized for their morphology, mechanical properties, thermal degradation, hardness and weathering resistance. In order to improve environment stability and abrasion resistance of the NR/MHSP composites, EPDM/MHSP were prepared and laminated on NR/MHSP and vulcanized. The laminated vulcanizated composites possessed the properties follow Thai Industrial Standard for rubber paving blocks could be obtained. The results also showed that NR filled with 50 phr of MHSP in NR and in EPDM gave the properties follow the standard and has high potential for outdoor rubber paving block.

1. Introduction
Vulcanized NR has been known for its excellent properties and extensively used in tire applications. In the rubber industry fillers are added to improve rubber compounds. The fillers are aimed either to improve electrical properties, mechanical properties, thermal resistance or cost reduction. Carbon black is the most important reinforcing filler used in the rubber industry. However, it causes pollution and also gives black colour to the rubber. With the awareness of environmentally global protection, biocomposites between rubber and natural based filler have been interested [3]. Natural fillers can be varied from natural fibres to particulate producing natural fibre/NR and particulate/NR composites. The application of rubber biocomposites have been ranged from aerospace, automotive to building
materials. Rubber tile for paver block or pavement block could be useful for ageing society due to its great properties such as shock absorption preventing elder age injuries. Floor and walkway tile, sport and play surface and sheet rubber are manufactured using crumb rubber particle using polyurethane as binder [4]. In this research we compound NR with mahogany fruit shell powder (MHSP), as natural filler, in order to obtained required hardness of vulcanized NR follow rubber tile standard and applied for rubber pavement block. Mahogany fruit shell contains 75.61% holocellulose and 13.54% lignin. The rubber block will be laminated with MHSP/EPDM vulcanizate in order to protect MHSP/NR composite from environmentally degradation.

2. Experimental
Materials: NR STR5L and EPDM 4725P containing ethylene and ENB of 70 and 4.9 % by mass respectively, were obtained from CHAREON TUT Co. Ltd. Thailand. Curing additives which are ZnO, stearic acid, MBT, DPG, CTP, 6PPD and sulphur were kindly supplied by Chemical Innovation group, Thailand. MHSP/rubber composites compound was prepared using efficient vulcanization system. Ground samples of Mahogany were separated according to particle size using a sieve shaker. Mesh size of sieves was 0.3, 0.15, 0.106 and 0.075 mm. Each sample was placed in the top sieve with the largest mesh and shaken for 5 min at an amplitude setting of 2 mm, disassembled and stirred lightly, then shaken for additional 5 min. The content of MHSP with particle size was varied from 0, 10, 20, 30, 40 and 50 phr. For EPDM was compound using efficient vulcanization and filled with MHSP at the same content as former rubber composites. Rubber compounds were tested using MDR, Gotech2000, Taiwan, at 160 ºC for cure characteristic parameters. Vulcanization was carried out in compression moulding at 160 ºC under pressure of 1500 psi. The vulcanized rubber composites were evaluated for their tensile properties and hardness using UTM, Instron model 5965, USA. Weathering resistance of the composites were evaluated according to ASTM G154 and the degree of colour fastness was measured using colour meter. The highest hardness MHSP/NR was selected and laminated with EPDM composite with the laminated thickness of 1-2 mm to obtain rubber tile 4 mm thick according to Thai Industrial Standard [5]. Hardness, Weathering resistance was also evaluated.

3. Results and discussion
Cure characteristic of MHSP/NR and MHSP/EPDM are presents in Figure 1 (a)-(f).

![Figure 1](image-url) Scorch time, delta torque and maximum cure rate of MHSP/NR (a)-(c) and MHSP/EPDM (d)-(f).
The results show that the addition of MHSP into both rubbers can accelerate vulcanization reaction, as can be seen by the shortening of scorch time and maximum cure rate index. This should be due to the organic substance contained in mahogany fruit shell such as fatty acid [6] which can act as co activator in sulfur vulcanization. In term of mechanical properties, MHSP could act as reinforcement in NR and EPDM and resulted in the increased in secant modulus as shown in Figure 2(a) and (d). In order to obtain hardness of rubber tile higher than 50 Shore A, according to Thai industrial standard, the content of MHSP added into rubbers should be higher than 40 phr, as shown in Figure 2 (c) and (d).

**Figure 2** Secant modulus at 100 % strain, elongation at break and hardness of MHSP/NR (a)-(c) and MHSP/EPDM (d)-(f).

Environmental resistance of MHSP/rubber composites were also examined and reported as degree of colour fastness depicted in Table 1. The results show that MHSP/NR composite with high MHSP content has lower degree of fastness than low MHSP content this should be due to the lignin content in MHSP. In fact, the degree of fastness should be higher than 4 and all composites have passed the standard of rubber tile.

**Table 1** Degree of colour fastness of rubber/Mahogany fruit shell powder composite after weathering test (environmental resistance).

| Formulation   | Degree of colour fastness | Formulation       | Degree of colour fastness |
|---------------|---------------------------|-------------------|---------------------------|
| NR            | 3                         | EPDM              | 1                         |
| NR/MHSP 10 phr| 4                         | EPDM/MHSP 10 phr  | 2                         |
| NR/MHSP 20 phr| 4                         | EPDM/MHSP 20 phr  | 3                         |
| NR/MHSP 30 phr| 4                         | EPDM/MHSP 30 phr  | (2-3)                     |
| NR/MHSP 40 phr| (3-4)                     | EPDM/MHSP 40 phr  | 3                         |
| NR/MHSP 50 phr| (3-4)                     | EPDM/MHSP 50 phr  | 3                         |

MHSP/NR composite and MHSP/EPDM composites contained MHSP 50 phr were selected to prepare laminated composite and evaluated for their hardness and degree of colour fastness. The thickness of EPDM composite laminate was varied from 1 to 2 mm. The hardness was found at 75 and
77 shore A respectively. Also, the degree of colour fastness was found at 3-4 in both 1 and 2 mm EPDM composite laminate thickness showing that both types of EPDM laminated MHSP/NR composite are qualified according to Thai Industrial standard for rubber tile. It is also useful to confirm that after weathering test hardness of both laminated rubber composite was not changed due to the environmental resistance of MHSP/EPDM composite. However, physical appearance was slightly changed due to the MHSP itself as shown in Figure 3. This colour change followed Thai industry standard for rubber tile as mentioned earlier.

Figure 3 Digital picture of MHSP/NR composite laminated with MHSP/EPDM composite the laminated thickness of 1 mm (a, a’) and 2 mm (b, b’) before (a, b) and after (a’, b’) weathering test.

4. Conclusions
In this research, rubber composites using mahogany fruit shell powder as reinforcing filler were successfully prepared. The composite with MHSP 50 phr possess required hardness for rubber tile, according to Thai Industry Standard. The composite was applied for rubber tile by laminate MHSP/EPDM on to MHSP/NR in order to protect MHSP/NR from environmental deterioration. The lamination thickness that was examined was 1 and 2 mm and found that hardness of both types of tile still follow the standard. In addition, the degree of colour fastness also agrees well with the standard. The results confirmed that the laminated composite are able to be used for pavement block particularly advantage for ageing society.

5. References
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Acknowledgement
The authors would like to thank the Department of Materials Science and Engineering, Faculty of Engineering and Industrial Technology, Silpakorn University for their support throughout their work.