PERFORMANCE OF BITUMEN IMPROVED WITH COCONUT SHELL ACTIVATED CARBON ADDITIVES

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Civil Engineering)

Faculty of Electrical Engineering
Universiti Teknologi Malaysia

JANUARY 2023

DEDICATION

This thesis is dedicated to my beloved father and mother,

Mamat Bin Embok and Minah Binti Mohamad,

My love,

Mohamad Fahmi Bin Pathil,

My beloved children,

Izz Danish, Izz Zara Sofia, Izz Armel Nufa and Izz Azfar,

All my brothers and sisters,

(Nizam, Zanariah, Nazri, Roslina, Zuliana, Nazaruddin and Hasimah)

That always inspires, loves and stands beside me.

ACKNOWLEDGEMENT

Praise and thanks to Allah S.W.T for his blessing, which enabled me to complete my PhD project. Firstly, I would like to express my sincere appreciation to my supervisor, Professor Ts Dr Mohd Rosli Bin Haini, for his encouragement, guidance, critics and friendship. I am also very thankful to my co-supervisor, Ts Dr Norhidayah Binti Abdul Hassan, for her guidance, advice and motivation. My special thanks go to Dr Mohd Idham, Dr Naqiuddin, Dr Ramadhansyah and Dr Zul Hanif. Without their continued support and interest, this thesis would not have been the same as presented here.

Special thanks are also extended to all technicians of Highway and Transportation Laboratory UTM, Mr Azri, Mr Azman, Mr Izwan, and Mr Sahak, for their time, help during the experimental laboratory work and valuable advice given throughout this project.

Last but not least, my fellow postgraduate student should also be recognised for their support. My sincere appreciation also extends to Dr Azah, Dr Nur Amira, Siti Nur Naqibah, Dr Izzul, Nizam, Dr Ibrahim, Dr Nurul Hidayah, and others who have assisted on various occasions. Their views and tips are helpful indeed. Finally, I am grateful to all my family members.

ABSTRACT

The agricultural industry alone generates a wide range of waste materials, such as shells, palm oil, husks, leaves, and fronds. Because of the growing waste generation, researchers have investigated alternative methods to recycle waste materials for bitumen modification to improve road materials and reduce solid waste pollution. While the utilisation of agricultural wastes for activated carbon production has been reported, there is limited study on the potential use of activated carbon from Coconut Shells (CS) in bitumen mixtures for road construction purposes. Therefore, this study investigates the feasibility of Coconut Shell Carbon (CSC) and Coconut Shell Activated Carbon (CSAC) as additives for road construction. The CSC and CSAC were used to modify 80/100 penetration grade bitumen (80/100PEN) under three work phases. In phase one, the CS waste was converted into CSC and CSAC, followed by the production and characterisation of the additives using the Laser Particle Size Analyser (LPSA) and Elemental Analyser (EA) for proximate and ultimate analysis respectively. CSC and CSAC were then mixed in 80/100PEN bitumen at 5% to 25% to produce the modified binder. The physical, chemical, and rheological properties of the modified binder containing CSC and CSAC were evaluated in the second phase under various ageing conditions (unaged, short- and long-term aged). The laboratory tests include Fourier Transform Infrared (FTIR), softening point, penetration, storage stability, viscosity, and Dynamic Shear Rheometer (DSR). The best modified binder was determined and used to develop the modified Hot Mixture Asphalt (HMA). For the third phase, the study focused on evaluating the mechanical properties of the modified HMA in terms of Marshall stability, resilient modulus, dynamic creep, indirect tensile strength, and rutting potential. The performance of the modified HMA was compared to the control of 80/100PEN HMA and 60/70PEN HMA. According to the results, both CSC and CSAC are harder, more corrosion-resistant and lower in emissions than that of the controlled mixtures. Furthermore, the chemical activation process of CS produces CSAC with a smaller microporous pore structure than CSC. When applied as additives to the bitumen mixture, the physical and rheological properties of the modified binder have been significantly improved, including stiffness and rutting resistance. The addition of 10% CSC and 15% CSAC per weight of 80/100PEN bitumen was the best proportion for the formulation of the modified binding with optimal properties. Compared to the controlled mixtures, the 5.2% CSAC improved by 81%, 46% and 13% in rutting, fatigue, and permanent deformation resistance, respectively. It can be concluded that CSAC can be used as an additive to change the properties of bitumen, improve performance, and promote the element of sustainability in road pavement.

ABSTRAK

Industri pertanian menghasilkan pelbagai jenis bahan buangan, seperti cengkerang, minyak sawit, sekam, daun dan pelepah. Memandangkan penjanaan sisa yang semakin meningkat, penyelidik telah menyiasat kaedah alternatif untuk mengitar semula bahan buangan bagi pengubahsuaian bitumen dalam proses menambah baik bahan jalan dan mengurangkan pencemaran sisa pepejal. Walaupun penggunaan sisa pertanian untuk pengeluaran karbon teraktif telah dilaporkan, terdapat kajian terhad mengenai potensi penggunaan karbon teraktif daripada Tempurung Kelapa (CS) dalam campuran bitumen untuk tujuan pembinaan jalan raya. Oleh itu, kajian ini menyiasat kebolehlaksanaan Karbon Tempurung Kelapa (CSC) dan Karbon Aktif Tempurung Kelapa (CSAC) sebagai pengubah suai bitumen untuk pembinaan jalan raya. CSC dan CSAC digunakan untuk mengubah suai bitumen gred penembusan 80/100 (80/100PEN) di bawah tiga fasa kerja. Dalam fasa satu, sisa CS ditukar kepada CSC dan CSAC, diikuti dengan pengeluaran dan pencirian bahan tambahan menggunakan Laser Particle Size Analyzer (LPSA) dan Elemental Analyzer (EA) untuk analisis proksimat dan muktamad. CSC dan CSAC kemudiannya dicampurkan dalam bitumen 80/100PEN pada 5% hingga 25% untuk menghasilkan pengikat yang diubah suai. Dalam fasa kedua, sifat fizikal, kimia dan reologi bagi pengikat diubah suai yang mengandungi CSC dan CSAC dinilai di bawah keadaan penuaan yang berbeza (tidak tua dan berumur jangka pendek dan panjang). Ujian makmal termasuk Fourier Transform Infrared (FTIR), titik lembut, penembusan, kestabilan penyimpanan, kelikatan, dan Dynamic Shear Rheometer (DSR). Pengikat ubah suai terbaik telah ditentukan dan digunakan untuk membangunkan Asfalt Campuran Panas (HMA) yang diubah suai. Untuk fasa ketiga, kajian tertumpu pada menilai sifat mekanikal HMA yang diubah suai dari segi kestabilan Marshall, modulus resilient, rayapan dinamik, kekuatan tegangan tidak langsung dan potensi rutting. Prestasi HMA yang diubah suai dibandingkan dengan kawalan 80/100PEN HMA dan 60/70PEN HMA. Mengikut keputusan, kedua-dua CSC dan CSAC adalah lebih keras, lebih tahan kakisan dan lebih rendah dalam pelepasan daripada campuran terkawal. Tambahan pula, proses pengaktifan kimia CS menghasilkan CSAC dengan struktur liang mikroporous yang lebih kecil daripada CSC. Apabila digunakan sebagai bahan tambahan kepada campuran bitumen, sifat fizikal dan reologi pengikat yang diubah suai telah dipertingkatkan dengan ketara, termasuk kekakuan dan rintangan alur. Penambahan 10% CSC dan 15% CSAC setiap berat bitumen 80/100PEN adalah perkadaran terbaik untuk perumusan pengikatan diubah suai dengan sifat optimum. Berbanding dengan campuran terkawal, 5.2% CSAC masing-masing meningkatkan peningkatan sebanyak 81 %, 46% dan 13% dalam rintangan terhadap aluran, daya ketahanan keretakan dan ubah bentuk kekal. Dapat disimpulkan bahawa CSAC boleh digunakan sebagai bahan tambahan untuk mengubah sifat bitumen dan meningkatkan prestasi serta menggalakkan elemen kelestarian dalam turapan jalan

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LIST OF ABBREVIATIONS

AASHTO	-	American Association of State Highway and Transportation Official
AC 14	-	Asphaltic concrete of nominal maximum aggregate size 14
AC-EFB	-	Activated Carbon Empty Fruit Bunch
ANOVA	-	Analysis of Variance
AP	-	Amorphous Polialfaolefin
ASTM	-	American Society for Testing and Material
ATR	-	Attennated Total Reflectance
BET	-	Brunauer–Emmett–Teller
BRHA	-	Black Rice Husk Ash
BT	-	Bentonite
C=O	-	Carbonyl band
CAGR	-	Compound Annual Growth Rate
CHNS	-	Carbon, hydrogen, nitrogen, and sulphur
CREaTE	-	Centre of Excellence for Engineering and Technology
CS	-	Coconut Shell
CSAC	-	Coconut Shell Activated Carbon
CSC	-	Coconut Shell Carbon
CSP	-	Coconut Shell Powder
CSS	-	Creep Strain Slope
DSA	-	Date Seed Ash
DSR	-	Dynamic Shear Rheometer
EA	-	Elemental Analyser
EFB	-	Empty Fruit Bunch
FTIR	-	Fourier Transform Infrared
HMA	-	Hot Mix Asphalt

ITS	-	Indirect Tensile Strength
JKR	-	Jabatan Kerja Raya
LPSA	-	Laser Particle Size Analyzer
MSCR	-	Multiple Stress Creep Recovery
MT	-	Million Tonnes
MT	-	Million Tonnes
PAV	-	Pressurised Ageing Vessel
PE	-	Polyolefin
PH	-	Potential of Hydrogen
POS	-	Palm Oil Ash
RHA	-	Rice Husk Ash
RTFO	-	Rolling Thin Film Oven
S=O	-	Sulfoxide band
SEM	-	Scanning Electron Microscopy
SHRP	-	Strategic Highway Research Program
TMD	-	Theoretical Maximum Density
UTHM	-	Universiti Teknologi Tun Hussein Onn Malaysia
UTM	-	Universal Testing Machine
VFB	-	Voids Filled with Bitumen
VMA	-	Voids in Mineral Aggregate
VTM	-	Voids in Total Mix
XRD	-	X-Ray Diffraction
XRF	-	X-Ray Fluorescence

LIST OF SYMBOLS

δ	-	The phase angle
D, d	-	Diameter
CO^2	-	Carbon Dioxide
E*	-	Dynamix Complex Modulus
G*	-	Complex Shear Modulus
T	-	Angular Frequency
r	-	Radius
G"	-	Storage Modulus
O^2	-	Oxygen
ε	-	Strain at 3600 cycles, mm
ε_{3600}	-	Strain at 3600 cycles
ε_{2000}	-	Strain at 2000 cycles
°C	-	Degree Celsius
M_R	-	Resilient modulus
St	-	Indirect tensile strength, kPa
E	-	Creep stiffness modulus, MPa
ζd	-	Maximum applied load (stress)
ζ	-	Applied stress, 300 kPa
E	-	Creep Stiffness Modulus, MPa
G_{mm}	-	Maximum specific gravity of the mixture
Gsb	-	Bulk specific gravity of aggregate
Gse	-	Effective specific gravity of aggregate

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Research Background

The 21st century is witnessing a change in the exploration of new sources of materials as the available natural resources are depleting at an unprecedented rate. Growing energy costs and global demand for oil encourage the development of alternative bindings, which can be modified or replaced by asphalt bindings using waste. While asphalt has been recycled and reused for road pavement works, more research is needed to maximise its performance, especially sustainability and durability.

Recently, emphasis on sustainability in road construction has been implemented in order to create healthier environments, optimize economic feasibility and ensure road safety. One of the strategies is to utilise agricultural waste. These cheap and abundant materials provide many mechanical and chemical advantages. In Malaysia, 95% of coconut growers are small farmers, with an average production of 6468 kg/hectare of coconuts in 2019. Between 2018 and 2019, coconut production increased to 495,531. MT to 527,729 MT (Jabatan Pertanian Semenanjung Malaysia, 2019; Nordin M. N, 2021). Figure 1.1 shows the global production of coconuts. Coconuts are grown in more than 94 countries and Malaysia is the world"s ten largest coconut producer (Sujatha, A., 2021). Examples of large-scale agricultural by-products include coconut shells, coconut clinkers, corn ash and rice ash. The application of agricultural waste in construction is likely to reduce global environmental pollution. In fact, industrial waste is used as an alternative to modified carbon in the construction process of road building mixes with bitumen and indirectly, it promotes the sustainability of the industrial environment.

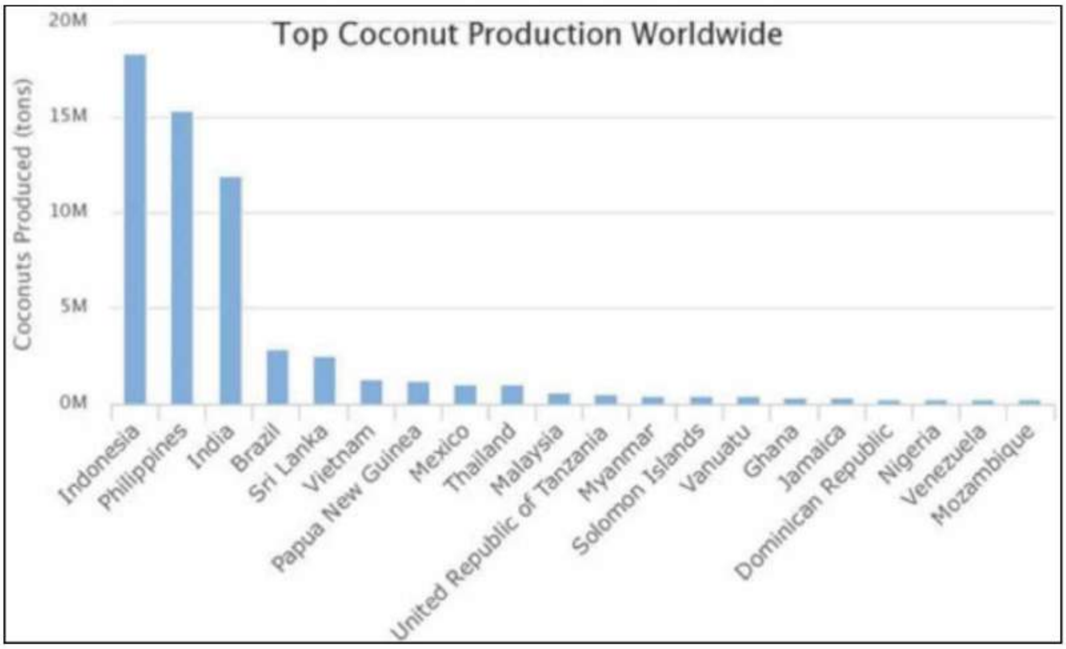

Figure 1.1 Coconut production worldwide (Sujatha.A et.al, 2021)

Another excellent material that has gained increasing demand to treat low concentrations of wastewater streams and harmful pollutants is activated carbon. Activated carbon also has a relatively high volatile content (above 50%). Volatile content is a substance that can be easily converted from solids or liquids to vapor. It is one of the most common parameters and part of the standard proximate analysis. Its contents include butane, isobutane, propan, hydrogen, carbon dioxide and oxygen.

The texture characteristics and surface properties of activated carbon depend on the raw material and the preparation method (Hidayu and Muda, 2016). Activated carbons, which can be prepared via the physical or chemical method. Considering the overwhelming global demand for activated carbon, biomass derivatives are seen as an interesting source of raw materials for activated carbon production due to their abundant supply and low price. In addition, it usually has a low inorganic content, which is a desirable property in activated carbon because carbon yields are generally low. The low amount of inorganic matter can affect the adsorption properties. The results of adsorption depend on the structural characteristics and surface chemistry of the adsorbent, the pH of the solution and the characteristics of inorganic pollutants.

1.2 Problem Statement

In the last 100 years, the pavement industry has experienced a significant change. The continuous demand for construction materials leads to a shortage of natural resources and an increase in waste. Additive is found to be the one method to improve workability and properties of the bituminous mixture. One of the sources of potential raw materials is biomass. Biomass can be converted through thermal and biochemical conversion and can further be developed into activated carbon. Many studies have shown that activated carbon attracts the attention of the whole world due to its high wealth, relatively low processing costs, and environmental-friendly nature. Recently, many studies have reported on the conversion of activated carbon from waste biomass, including the shells of date stone palm and coconut (Syairah et al., 2018; Shariff et al., 2016; Teh et al., 2015) and rice husk (Liu et al.,2016). Since environmental pollution is today the main problem, the need for activated carbon is increasing every day.

Research on the coconut shell as an additive in bitumen and aggregate replacement has attracted the attention of the researcher (Jaya et al., 2019; Abdullah et al., 2017). Many studies have proved that Coconut Shell (CS) can be integrated into the original mixture of bitumen. For example, (Buhari et al., 2016) have discovered the possibility and effect of using coconut shell powder (CSP) as a binding additive. In addition, Coconut Shell Coal (CSC) Ash is a derivative of Coconut Shell (CS) and is used as a bitumen moderator to improve the composition of the binder (Abdullah et al., 2017). However, innovation in utilization of activated carbon made of coconut shell is still uncommon and new in Malaysian construction industry. The addition of natural additives such as coconut shell activated carbon to asphalt mixtures is very rare. There is still a lack of research on the performance of activated carbon from coconut shell (CSAC). In particular, it were discovered in research on CSAC additives to be incorporated into the bitumen mixture.

Furthermore, many researchers have pointed out that agricultural products are used to produce activated carbon. It was also discovered that agricultural waste is renewable and relatively inexpensive as a precursor of active carbon (Yahya et al., 2015). Most current research on CSAC generally focuses on the process and sustainability of the materials, while the performance, properties and characteristics of CSAC are very limited. In CSAC study, (Das et al., 2015) focused on preparation of activated carbon from coconut shell and its characterization, while (Islam et al., 2015) studied the adsorption capacity of activated carbon synthesized from the shell of the coconut.

Issue of incorporating CSAC as an additive has also motivated the research, where it will focus more specifically on the bituminous mixture. Furthermore, the results of this residue are also crucial for improving the chemical, physical and rheological properties of the activated carbon in coconut shells and their potential applications as asphalt additives. The results are expected to also improve mechanical performance in road construction. Overall, this study is very significant to encourage researchers to conduct further studies on the use of CSAC in road construction.

1.3 Objective of Study

The main aim of the study is to investigate the feasibility of the waste of coconut shell (CS), which include coconut shell carbon (CSAC) and coconut shell activated carbon (CSAC) as additives for road construction. The goal of this study is achieved through the following research objectives:

i) To determine the characteristics and feasibility of CSAC as additives.
ii) To investigate the physical, chemical, and rheological properties of the bitumen binder with CSAC.
iii) To evaluate the performance of the Hot Mix Asphalt (HMA) containing CSAC.

1.4 Scope of Research

The scope of the study involves the addition of the conventional 80/100 penetration grade bitumen (80/100PEN) using 0.0075mm of CSC and CSAC, followed by the physical, chemical, and rheological analysis of the modified binder. Finally, the feasibility of the optimum binder was determined by incorporating it in the HMA and subjected to several performance tests. The CS waste was collected from different locations around the Skudai area in Johor, while CSAC was purchased from a factory in Perak. Asphalt concrete is used for asphalt mixtures for the entire study period, and the maximum aggregate size is 14 mm (AC14). In addition, the standard 80/100PEN bitumen was used to produce the control and mixture samples. The aggregates were collected from an Ulu Choh quarry in Pulai. Following the addition of CSC and CSAC, the chemical, physical, and performance characteristics of the mixture samples were compared with control characteristics. All laboratory tests were based on the American Society for Testing and Material (ASTM), JKR Specification, and the American Association of State Highway and Transportation Officials (ASSHTO).

1.5 Significance of Research

Based on the obtained results from this study, the significance of this research as in the following:

i) The finding would reveal new properties of mix 80/100PEN bitumen that might be comparable with the 60/70PEN. Moreover, it would be desirable to be utilised as a new binder. CSC and CSAC is often used as additive to improve the workability, increase the appropriate interval of service temperature and improved resistance to chemical aging.

ii) The unpredictable weather in Malaysia is one of the factors that can weaken the pavement structure. The use of CSAC as additives in bitumen mixture could improve the quality of road and pavement construction. However, some effort is needed to implement the new binder in road construction efficiently. The using CSAC additive would exhibit better rutting and fatigue resistance, also improved strength and durability than the control bitumen.

iii) In terms of addressing the environmental concerns, the exploitation of waste materials from CS as activated carbon in bitumen modification would promote a recycling-based society and arrest the fast-declining natural resources. Ultimately, such efforts would reduce waste materials and potentially reduce pollution.

iv) Using CSC and CSAC as additives would provide an alternative method to utilise the alarming increase of waste materials and promote sustainability. Sustainability in construction is one of the efforts to create healthier environments and reduce environmental impacts that developed countries are pursuing.

v) The nature of agricultural waste has many advantages, including high specific strength and modulus, low density, biodegradability, absence of health hazards. This advantage is advantageous to the construction industry, as it improves the quality of composites and is cheap compared to other construction materials.

1.6 Thesis Outline

This thesis consists of five chapters, and the details are as follows:

i) Chapter 1 discusses the background of the study, an overview of the problem statement, establishes the research objective and states the significance of this study.

ii) Chapter 2 presents a comprehensive literature review in related areas of this study. This covers the scope of CS, CSC, CSAC, and the properties of the modified binder. Information about rutting damage and fatigue cracking is also included.

iii) Chapter 3 explains the process and methods used in this study. There are three phases of the operational framework comprising the material production and characterisation, method of preparation of the materials, the equipment, laboratory testing procedures, and the applied standard specification.

iv) Chapter 4 presents the laboratory testing results. The characteristic of CSC and CSAC, the physical, chemical, and rheological properties, and the performance of the new binder and bitumen mixture are all presented and discussed.

v) Chapter 5 concludes the finding, and a list of recommendations for subsequent research in the future is provided.

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