CHARACTERIZATION OF HYDROPHOBIC-TREATED RECYCLED PAPER MILL SLUDGE IN BITUMINOUS MATERIALS

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ABSTRAK

Peningkatan penghasilan sisa pepejal dan aktiviti pelupusan akibat perkembangan kawasan bandar yang semakin berkembang telah mengehadkan ruang pelupusan. Industri turapan asfalt mengambil peluang ini untuk memanfaatkan sisa pepejal sebagai bahan pengubahsuai dalam campuran bitumen sambil mempraktikkan alternatif yang mampan dalam teknologi turapan asfalt. Penyelidikan ini mengimplementasikan Enapcemar Kitar Semula dari Kilang Kertas (RPMS) sebagai bahan tambahan sisa pepejal bagi menggantikan separa pengisi mineral dalam campuran asfalt. RPMS mentah memerlukan pengeringan oven selama 24 jam untuk menyejatkan kandungan air sepenuhnya sebelum pengurangan saiz dengan kaedah pengisaran RPMS kering. RPMS yang tertahan di pengayak berukuran 0.075mm dan dulang penahan (0.075mm – 0.00mm) yang tidak dimodifikasi diperoleh melalui proses penggilingan dan pengayakan di mana RPMS yang melepasi dan tertahan pada dulang ayak berukuran 75µm akan dikumpulkan dan disimpan di dalam bekas kedap udara untuk kegunaan penyelidikan. Penggredan zarah RPMS dinilai menggunakan ujian taburan saiz zarah. Seterusnya, RPMS dirawat melalui kaedah etanol yang melibatkan esterifikasi etil ester dengan menggunakan 7-ml sisa minyak masak (WCO) dan 50-ml etanol. Kaedah rawatan ini menghasilkan RPMS yang lebih hidrofobik apabila pekali hidrofilik lebih rendah dan sudut sentuhan air lebih tinggi daripada RPMS yang tidak dimodifikasikan. Hasil dari ujian mikroskop elektron imbasan (SEM), ia menggambarkan RPMS yang telah diubahsuai mengandungi komponen yang kurang berserabut, tekstur permukaan kasar dan berpori, pengumpalan zarah dan bentuk yang seragam. RPMS ditambah ke dalam campuran asfalt dengan kandungan 0.5% dan 1.0% berdasarkan berat agregat. Dalam penyelidikan ini, pengikat asfalt bergred 60/70 dan agregat granit telah digunakan. Campuran asfalt dicampurkan dan dipadatkan masing-masing pada suhu 160℃ dan 150℃. Kedua-dua campuran asfalt yang dimodifikasi telah mencapai peningkatan dalam kandungan pengikat optimum (OBC). Kekuatan penjerapan RPMS terubahsuai adalah lebih tinggi berbanding pengisi biasa seperti batu kapur dan granit. Sifat mekanikal seperti kestabilan, aliran, dan keutuhan campuran asfalt diubahsuai dinilai melalui ujian kestabilan Marshall. Hasil kajian menunjukkan bahawa ketiga-tiga sifat mekanikal tersebut menurun apabila RPMS terubahsuai diimplementasikan. Walaupun begitu, keputusan tersebut masih berada di dalam jangkauan keperluan Jabatan Kerja Raya (JKR) dan ini menunjukkan bahawa penggunaan RPMS adalah praktikal. Prosedur larut lesap pencirian ketoksikan (TCLP) membuktikan bahawa hanya Zink (Zn), Tembaga (Cu) dan Kadmium (Cd) sahaja yang telah dikesan melampaui had peraturan yang selamat di Malaysia. Walau bagaimanapun, Zn dan Cu tidak tersenarai di dalam bahan merbahaya yang terlarang dan menjadi komponen asas bagi makhluk hidup dan mikroorganisma. Penyelidikan ini mengimplikasikan bahawa pengalikasian RPMS dalam campuran asfalt merupakan penyediaan terhadap teknologi mampan dan penggantian pelupusan yang lebih hijau dalam industri asfalt.

ABSTRACT

The increasing amount of solid waste generated and disposal activities due to massive urbanisation has led to limited landfill space. Asphalt pavement industry has seized this opportunity to transform the solid wastes as modifiers into bituminous mixtures while practising a sustainable alternative in asphalt pavement technology. This research implements Recycled Paper Mill Sludge (RPMS) as a solid waste additive to partially replace the mineral filler in asphalt mixture. The raw RPMS required a 24-hour oven drying to completely eliminate its water content, prior to the reduction of size by granulating and grinding the dried RPMS. The 0.075mm and pan size unmodified RPMS are obtained by sieving process in which those retained on 75µm and pan size sieve are collected and kept in an air-tight container for the research usage. The particles of RPMS are determined by particle size distribution test. RPMS is modified by ethanol method which involving an esterification of ethyl esters that utilizing 7ml of waste cooking oil (WCO) and 50ml of ethanol. This treatment method yielded modified RPMS with improved hydrophobicity which resulted in a lower hydrophilic coefficient and higher water contact angle than the unmodified RPMS. From Scanning Electron Microscope (SEM), it illustrated the modified RPMS with less fibrous components, rough and porous surface texture, agglomeration of particles and regular shapes. The RPMS is incorporated into asphalt mixtures at 0.5% and 1.0% of the weight of aggregates. In this research, asphalt binder of 60/70 and granite aggregates are used. The asphalt mixture is mixed and compacted at temperatures of 160°C and 150°C respectively. Both modified asphalt mixtures achieved an increased optimum binder content (OBC). The adsorption strength is proved to be stronger for modified RPMS compared to common fillers such as limestone and granite dust from baghouse. Mechanical properties for instances stability, flow and stiffness of modified asphalt mixtures are determined from Marshall Stability test. The results showed that the three properties decreased with the incorporation of modified RPMS. Despite that, the results are within the Public Work Department (PWD) requirement range and hence the incorporation of RPMS is applicable. Toxicity Characterization Leaching Procedure (TCLP) has proven that only Zinc (Zn), Copper (Cu) and Cadmium (Cd) has found to be exceeded the safe regulatory limits in Malaysia. However, Zn and Cu are excluded from the list of restricted hazardous substances and being the fundamental components for living things and microorganisms. This research implies the implementation of RPMS in asphalt mixture is providing a sustainable technology and greener disposal replacement in asphalt paving industry.

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LIST OF SYMBOLS

Al	Aluminium
As	Arsenic
Cd	Cadmium
$CaCO_3$	Calcium Carbonate
Cr	Chromium
C_u	Coefficient of Continuity
Cu	Copper
Cn	Cyanide
V_{fa}	Effective Filler Volume
φ_e	Effective Particle Concentration
C_2H_6O	Ethanol
η	Hydrophilic Coefficient
OH-	Hydroxyl group
Fe	Iron
Pb	Lead
Mn	Manganese
M_b	Mass of binder
M_f	Mass of Filler
φ_m	Maximum Concentration Without Binder
Hg	Mercury
CH_3OH	Methanol
Ni	Nickel
φ	Particle Concentration
K_2CO_3	Potassium Carbonate

KOH	Potassium Hydroxide
C_2H_5NaO	Sodium Ethoxide
NaOH	Sodium Hydroxide
CH_3ONa	Sodium Methoxide
G_{sb}	Specific Gravity of Binder
G_{sf}	Specific Gravity of Filler
A	Specific Surface Area
Sn	Tin
$Sn(Oct)_2$	Tin (II) ethyl-hexanoate
V_f	Volume of Filler
V_{bfree}	Volume of Free Bitumen
Zn	Zinc

LIST OF ABBREVIATIONS

ABS	Acrylonitrile Butadiene Styrene (ABS)
AC	Asphalt Concrete
ACV	Aggregate Crushing Test
ATRP	Atom Transfer Radical Polymerization
BGA	Bagasse Ash
C&D	Construction and Demolition
CAN	Ceric Ammonium Nitrate
CFA	Coal Fly Ash
CME	Canola Oil Fatty Acid Methyl Ester
CNCs	Cellulose Nanocrystals
CRMB	Crumb Rubber Modified Binder
CW	Ceramic Waste
DPM	Department of Urban Planning and Municipalities
EBA	Ethylene-Butyl Acrylate
EI	Elongation Index
ESP	Eggshell Powder
EVA	Ethylene-Vinyl-Acetate
FAEEs	Fatty Acid Ethyl Esters
FAME	Fatty Acid Methyl Ester
FI	Flakiness Index
FRP	Free Radical Polymerization
GD	Diglyceride
GLY	Glycerol
GTM	Gyratory Testing Machine

HC	Hydrophilic Coefficient
HDPE	High-Density Polyethylene
HIPS	High Impact Polystryrene
HL	Hydrated Lime
HMA	Hot Mix Asphalt
ITS	Indirect Tensile Strength
ITSR	Indirect Tensile Strength Ratio
LAAV	Los Angeles Abrasion Test
LDPE	Low-Density Polyethylene
LLDPE	Linear Low-Density Polyethylene
MBT	Mechanical Biological Treatment
MCC	Microcrystalline Cellulose
MCC-PP	Microcrystalline Cellulose – Polypropylene
MeOH	Methanol
MG	Monoglyceride
MPOB	Malaysian Palm Oil Board
MSW	Municipal Solid Waste
NA	Natural Aggregate
OBC	Optimum Binder Content
OGFC	Open-graded Asphalt Friction Course
PBR	Polybutadiene Rubbers
PCL	Poly(e-Caprolactone)
PET	Polyethylene Terephthalate
PMMA	Poly (Methyl Methacrylate)
POFA	Palm Oil Fuel Ash
PP	Polypropylene
PPA	Polyphosphoric Acid

PVC	Polyvinyl Chloride
PWD	Public Works Department of Malaysia
RAP	Reclaimed Asphalt Pavement
RCA	Recycled Concrete Aggregate
RDRP	Reversible-Deactivation Radical Polymerization
RHA	Rice Husk Ash
ROP	Ring-Opening Polymerization
RPM	Revolutions Per Minute
RPMS	Recycled Paper Mill Sludge
RV	Rigden Voids Test
SBR	Styrene-Butadiene Rubber
SBS	Styrene-Butadiene-Styrene
SCBA	Sugarcane Bagasse Ash
SCG	Spent Coffee Ground Oil
SEBS	Styrene-Ethylene/Butylene-Styrene
SEM	Scanning Electron Microscopy
SFEEs	Sunflower Oil Fatty Acid Esters
SGC	Servopac Gyratory Compactor
SIS	Styrene-Isoprene-Styrene
SMA	Stone Matrix Asphalt
SS	Steel Slag
TCLP	Toxicity Characteristic Leaching Procedure
TG	Triglyceride
USEPA	United State Environmental Protection Agency
UTM	Universal Testing Machine
VFB	Voids in Aggregates Filled by Bitumen
VMA	Voids in Mineral Aggregates

WCA	Water Contact Angle
WCO	Waste Cooking Oil
WCO-SFEEs	Waste Cooking Oil Fatty Acid Ethyl Esters
WI	Workability Index
WMA	Warm Mix Asphalt
WTS	Wastewater Treatment Sludge

CHAPTER 1

INTRODUCTION

1.1 Background

Alternative approaches in recycling solid waste materials have gained attention among industry players and authorities in recent decades, owing to limited waste disposal space and stringent environmental laws. This is due to expanded production, resulting in vast amount of solid waste generated from industrial sectors, agricultural activities, as well as municipal and domestic waste. According to statistical data, there are 19,000 tons of solid wastes produced daily, with only 5% of the recycling rate in 2005. In 2006, the amount was escalated to 38,200 tons per day with 17.5% the recycling rate (Malaysia: Toward A Sustainable Waste Management, 2020). In general, this issue can be tackled by retrieving materials and energy from waste products and incorporating it into asphalt pavement industries to reduce the exploitation of natural resources and landfill usage (Rahman et al., 2020). By grabbing this opportunity, the asphalt pavement industry has become a field of interest for implementing solid wastes as modifiers into bituminous mixtures (Barbuta et al., 2015).

Asphaltic concrete typically consists of bitumen, various sizes of aggregates, and filler minerals. Approximately 90% of the mixtures constituted of aggregates, whereas fillers usually corresponding to 3% to 14% of the total aggregate by weight in the entire mixture irrespective of the type of asphalt mixtures (Wasilewska, 2017). The filler is referred to as the inert, finest portion of aggregate that can pass through a sieve size of 0.075mm in the United States, India, Malaysia and 0.063mm in Europe (Choudhary et al., 2020). Mineral filler plays a crucial role in filling the voids between coarse aggregates, strengthening the asphalt mixture, and reducing optimum binder content (Nathem, 2013).

Likewise, the significant amount of sludge generated from paper mills industries also causing environmental consequences. Jang et al., (2018) stated that sludge is mostly disposed by landfill or ocean disposal which in turns lead to secondary environmental pollution issues and it is usually disposed to landfills at a high cost. These disposal methods are not reliable as they might be causing ground water contamination and overuse the landfill capacity (Caputo and Pelagagge, 2001). As a result, this waste known as recycled paper mill sludge (RPMS) has also been utilized as a filler portion for the asphalt mixture productions. Recycled paper mill sludge is composed of fibrous recycled paper, chemical additives, organic matter originated from cellulose fiber (from wood or recycled paper), and inorganic compound (mainly consist of calcium carbonate, kaolinite, and talc). It is yielded from the paper-making industries as the final generated waste after several phases such as sorting, pulping, screening, cleaning, deinking, refining, color stripping, and bleaching processes (Abdullah et al., 2015).

In this study, hydrophobic-treated RPMS will be used as the asphalt modifier. Hydrophobic indicates the surfaces and structure of molecules having the property of repelling water. The non-polar molecules clumped together and farther away from water, thus inducing the hydrophobic effect (Chew et al., 2020). The hydrophobic-treated RPMS resulted in the water-repellent surface that reduces water or dissolved substances absorption, which could affect any material structural integrity. For this reason, hydrophobic treatment can enhance the resistance to moisture damage (Vries, 1997).

The establishment of this research is mainly to characterize the hydrophobic-treated RPMS in bituminous materials. The treated RPMS will undergo laboratory assessments to determine its physical and mechanical properties. Furthermore, hydrophobic treatment’s suitability and effectiveness to the RPMS by using canola oil or waste cooking oil with ethanol extracted through the esterification process have also been evaluated.

1.2 Problem Statement

In past decades, Malaysia has been experiencing phenomenal growth in population, urbanization, and industrialization. This rapid growth is reflected in the significant amount of solid wastes (Fauziah and Agamuthu, 2012). Malaysians generate 38,000 tons of solid waste per day from households, industry, commercial, and institutional sources, with the amount increasing at a 0.5% annual rate (Ministry of Energy Green Technology and Water, 2017). In general, Malaysia has reached a critical level in solid wastes generation, especially in terms of amount and composition. The accumulating amount of waste generated had caused a scarcity of available landfill capacity as it is the primary disposal method (Chew et al., 2020). Therefore, this research is one of the initiatives to solve the world's landfills challenge by recycling paper mill sludge as an asphalt modifier. It is viable and practical in lowering the cost of managing the waste and minimizing the depletion of natural resources concurrently (Barbuta et al., 2015).

However, the hydrophilic properties of RPMS can cause premature damage to the asphalt mixtures due to moisture-induced damage mechanisms. It is evaluated that asphalt mixture with incorporated RPMS has a lower indirect tensile strength. This indicates the mixture with RPMS has poor water resistance due to the presence of water-soluble mineralogical compounds namely anhydrite, halite, sylvine, quartz and periclase (Chew et al., 2020). Moisture damage or moisture-induced damage can significantly lower the pavement's strength and durability, especially for pavement with tropics. The moisture within the macropores and existing cracks percolated and impaired the pavement, leading to high moisture susceptibility during cyclic loading moisture damage acceleration (Yilmaz and Sargin, 2012). The presence of moisture has worsened the existing pavement distresses indirectly and impacted the short-term and long-term pavement performance. Permanent deformation such as cracking, rutting, upheaval, ravelling, corrugations, and shoving is expedited due to excessive moisture (Miller and Bellinger, 2003). Hence, it is crucial to develop mitigation actions to improve the pavement and prolonged their service life.

A longer life expectancy asphalt mixture is notable for sustainable consumption and production (SCP), stated in the eleventh Malaysia plan by Economic Planning Unit (2015). Pavement with a short lifespan triggers environmental and economic impact as it requires more maintenance and rehabilitation activities. By this concern, efficient use of natural resources such as better curing materials usages and practices should be adopted. Thereby, the asphalt modifier RPMS is required to undergo prerequisite treatment method by introducing hydrophobic treatment with hydrophobic agents such as canola oil extracted through esterification.

In short, laboratory work is essential to determine the suitability and effectiveness of the prerequisite treatment method to the RPMS to improve its hydrophilic properties. The optimum binder content required by the asphalt mixtures incorporated with pre-treated RPMS will be evaluated thoroughly and justify with the adsorption strength of treated fillers. Lastly, the performance of asphalt mixture with RPMS as asphalt modifier or additive will be discussed based on the mechanical properties that evaluated by Marshall Stability test.

1.3 Objective

The aim of this research is to characterize the performance of asphalt mixtures incorporating hydrophobic-treated recycled paper mill sludge to promote a green alternative for solid waste disposal and sustainable asphalt pavement. The specific objectives of this study are:

1. To assess the suitability and effectiveness of prerequisite hydrophobic treatment that applied to the RPMS.
2. To determine the optimum binder content (OBC) of asphalt mixture with treated RPMS as a partial replacement of mineral fillers and justify with its adsorption strength.
3. To evaluate the mechanical performances of asphalt mixture prepared with the hydrophobic-treated RPMS as modifier or additive in terms of stability, flow and stiffness in accordance with standard specifications.

1.4 Scope of Work

This research intends to characterize the physical and mechanical properties of asphalt mixtures incorporating hydrophobic-treated RPMS. This study begins with the preparation of raw material which consists of granite as aggregate, a binder with penetration grade of 60/70, and RPMS which undergoes prerequisite treatment. The grounded RPMS that is formerly treated with canola oil and waste cooking oil to attain hydrophobic properties is subjected to several property tests to determine its surface characteristics and composition mineralogy as well as physio-mechanical properties. The efficiency of the hydrophobic treatment will be evaluated based on the hydrophilic coefficient, water contact angle and its surface morphology via Scanning Electron Microscopy (SEM). The basic properties of granite aggregates and asphalt binder adopted for the experimental work will also be determined. Then, it is followed by the preparation of asphalt mixtures via Marshall method. Marshall Mix Design determines the optimum binder content (OBC) for modified asphalt mixture as well as its mechanical properties in terms of stability, flow and stiffness. Lastly, the adsorption strength as well as Toxicity Characterization Leaching Procedure (TCLP) of RPMS will be evaluated.

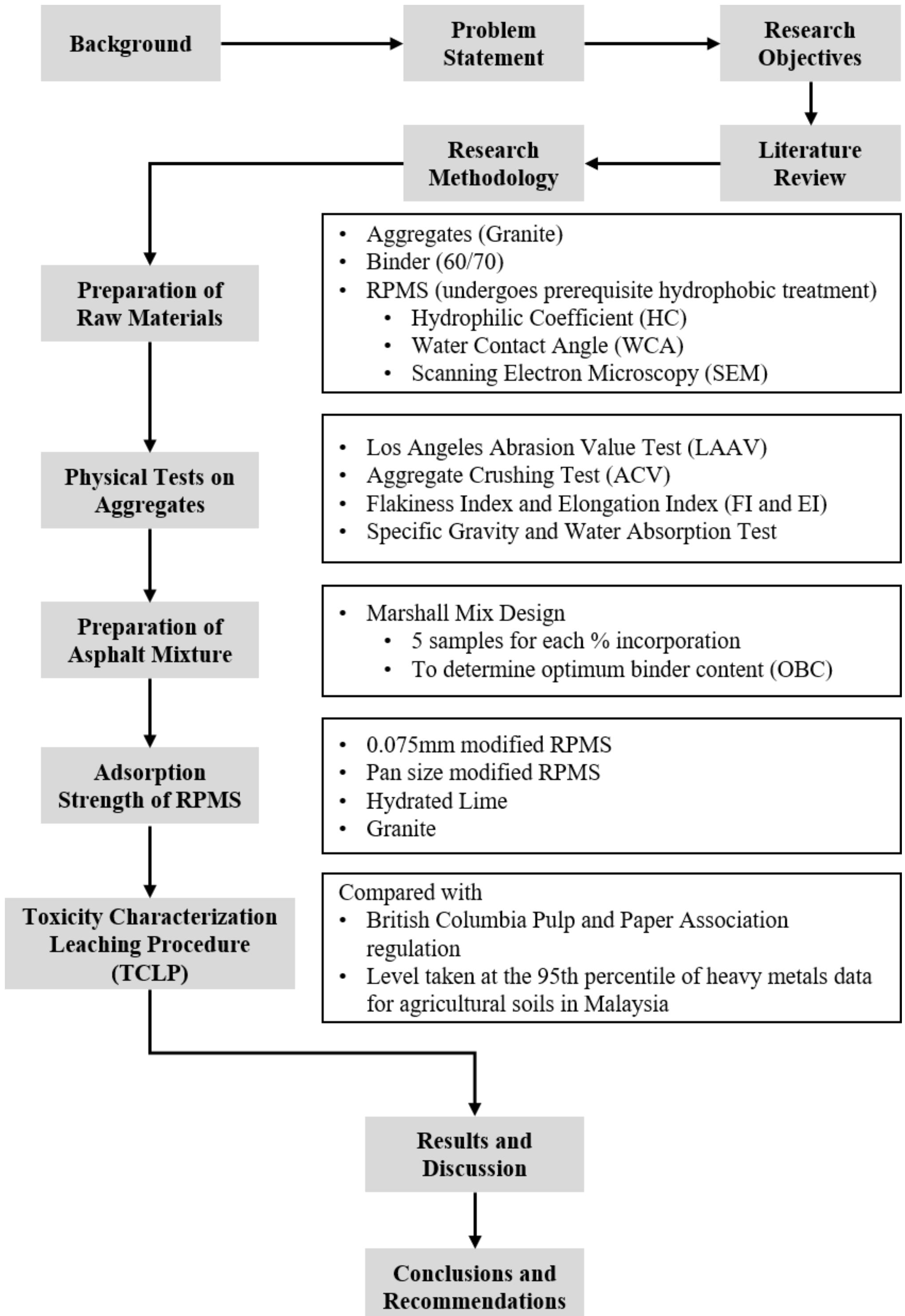

Figure 1.1: Workflow Planning for This Research

1.5 Significance of Study

In recent years, an increasing number of organizations, industries, and researchers have emphasized incorporating sustainability principles into asphalt pavement by recycling and reusing solid waste materials. This alters substantial savings in aggregate resources and demand for asphalt binders. Instead of primary or virgin materials, the utilization of secondary or recycled materials is more effective in alleviating landfill pressures while cultivating sustainable construction practices (Huang et al., 2007). As landfill capacity is getting scarce, adopting RPMS as an asphalt modifier is a practical approach that can minimize the amount of waste disposal and the disposal cost.

The pavement incorporated with recycled materials as asphalt modifiers or additive has proven to be equal to those prepared with raw materials in quality. Pavement distress is highly concerned by private or government agencies due to the tremendous budget requirement for maintenance and restoration of asphalt pavements. By this, pre-treated RPMS for its hydrophilic properties contributing to moisture damage can be adopted into asphalt pavement in practicing sustainable pavement production. The research aims to enhance the durability and strength of asphalt pavement that is added with RPMS as an asphalt modifier by introducing hydrophobic treatment. Improved pavement technology contributes to significant cost savings, efficient waste management, and increased economic feasibility while paving the way for long-term green asphalt pavement. (Chew et al., 2018).

1.6 Organization of Dissertation

This dissertation illustrates the introduction of research, literature review, materials used, and methodology that applied for this study. Results from tests, study findings, discussions, conclusions, and recommendations based on the conducted laboratory experimental works are also presented. There are five chapters in this dissertation, including Introduction, Literature Review, Research Methodology, Results and Discussions, and Conclusion and Recommendations.

Chapter One discussed the background for the research study, problem statement that identified the problem in the current environmental issues and technology, objectives to be achieved in this study, the scope of work, and study significance.

Chapter Two involved a collection of literature reviews consisting of past research findings relating to the implementation of solid waste into asphalt mixture, properties of recycled paper mill sludge (RPMS) that added as asphalt modifier into bituminous materials, and the effect of various hydrophobic treatments towards the bitumen.

Chapter Three described the materials used and methods of preparation to accomplish the research objectives. The properties tests that will be conducted in evaluating the characteristics of hydrophobic-treated RPMS, physical and mechanical properties of asphalt mixture will also be discussed in this chapter.

Chapter Four presented the results obtained from the tests conducted and discussion based on the collected laboratory data. The results of treatment method tests for RPMS and characterization for overall asphalt mixture is being analysed. It has also been discussed in detail and included in this chapter.

Chapter Five concluded the discussion that had been determined throughout the entire research study. Recommendations are also suggested for further research work. The objectives have been evaluated once again at the end of the research study.

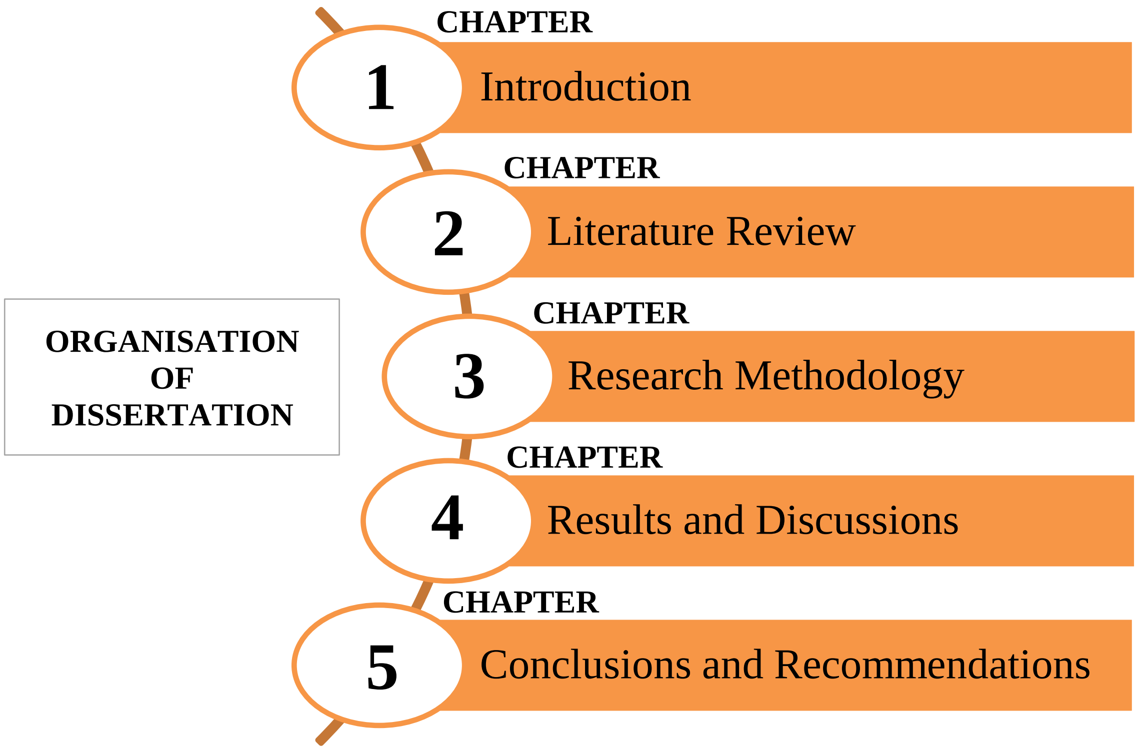

Figure 1.2: Contents of Dissertation

CHAPTER 2

LITERATURE REVIEW

2.0 Introduction

Asphalt binders are defined as viscoelastic materials that are strongly affected by the service temperature and type of load that is applied to it, such as traffic load. It shows ductile behaviour and is susceptible to the service temperature and plastic deformations (Brasileiro et al., 2019). Therefore, to reduce the impact of external factors on the strength of asphalt mixtures, the mixture properties can be enhanced by modifying them with either commercially available additives or additives from waste materials. The modification offers an opportunity to optimize the performance of asphalt mixtures and improve the deficient properties in relative to current standards. It tends to obtain asphaltic concrete with enhanced qualities such as adequate resistance towards rutting, thermal cracking, aging, permanent deformation, stripping, moisture damage, etc. Various additive materials, including fine fillers, aggregates, fibers, polymers, etc., are used to improve the asphalt concrete properties.

During the last few decades, an increasing number of pavement engineers or researchers began to focus on asphalt modification with waste materials (El-Badawy et al., 2019). Accelerated urbanization and civilization have produced vast amounts of waste, leading to severe disposal issues and an impact on the environment. Asphalt pavement industries have transformed the solid waste materials from the landfill into asphalt in mineral filler, aggregate or fiber asphalt modifiers. There is a significant amount of waste materials generated from various sources and can be classified into few categories, such as (a) household wastes (Azahar et al., 2016; Leng et al., 2018; Shafabakhsh et al., 2014; Mahssin et al., 2021; Choudhary J. et al., 2020), (b) agricultural wastes (Hainin et al., 2014, Mistry et al., 2019, Arabani et al., 2020, Ting et al., 2016), (c) Industrial wastes (Mashaan et al., 2014, Woszuk et al., 2019, Choudhary et al., 2019, Tao et al., 2019, Ali Jattak et al., 2019, Olugbenga, 2019) in this research study.

This chapter establishes a brief understanding of the commercial modifier and waste materials as alternative modifiers in asphalt mixtures, wastes sludge or materials that have been implemented worldwide. The performance evaluation methods to ensure the mixture develops sufficient strength in terms of physical and mechanical performances of the bituminous mix has also been reviewed and discussed.

2.1 Asphalt Modifications

2.1.1 Applications of Commercial Modifiers

In general, modifiers or additives are added to strengthen the asphaltic concrete. Asphalt mixtures modification can be done by using commercial modifiers or additives such as polymers, filler, chemical modifiers, antioxidants, antistripping agents, and stiffening agents (Daly, 2017).

Among the modifiers, polymer modification of bitumen is one of the most widely applied method. The earliest patent of modification that involved natural and synthetic polymers is recorded in 1843 (Thompson and Hoiberg, 1979) and then continue to be utilized by those developed and developing countries. Polymers can be divided into two categories named as elastomers and plastomers. Elastomers are defined as a class of polymeric material with high elastic properties which can resist permanent deformation. It includes styrene-butadiene rubber (SBR), styrene-butadiene-styrene (SBS) (Airey, 2003; Airey, 2004; Attaelmanan et al., 2011 Sengoz et al., 2008), styrene-isoprene-styrene (SIS) (Zhang and Zhang, 2018), styrene-ethylene/butylene-styrene (SEBS) (Polacco et al., 2006; Zapién-Castillo et al., 2016) and polybutadiene rubber (PBR) (Mansourkhaki et al., 2020). The SBS is the most frequently used modifier as it has effectively improved the performance of asphalt pavement (Daly, 2017).

Plastomers such as ethylene-vinyl-acetate (EVA) (Bulatovič et al., 2013), ethylene-butyl acrylate (EBA), polypropylene (PP) (Al-Hadidy et al., 2009), polyolefins such as high-density polyethylene (HDPE), low-density polyethylene (LDPE) and linear low-density polyethylene (LLDPE) have also been adopted as modifier in bitumen modification. These polymers effectively enhance the properties of bitumen in terms of temperature susceptibility, rutting, and cracking resistance (Hassanpour-Kasanagh et al., 2020; Yildirim, 2007).

Besides, filler mineral is a fundamental part in bituminous material, which impacts the strength of asphalt pavement. Hydrated lime (HL) has been the most popular filler for years (Shafiei et al., 2014; Kakade et al., 2016) to be applied in bitumen modification to improve stripping resistance against moisture damage. According to Diab et al., (2012), hydrated lime can be incorporated by using wet (dry HL with wet aggregates), dry (dry HL powder with dry aggregates) and slurry method (dry HL with treated aggregates). In 2010, Lee et al., (2010) has conducted a research to characterize the effect of HL on dynamic modulus and it resulted in an increased dynamic modulus in HMA. Kakade et al., (2018) has clearly reported that the addition of HL in bituminous mixed greatly reduced the potential rutting as well as the rut depth (Shafiei et al., 2014).

Stiffening agents such as polyphosphoric acid (PPA) and gilsonite are added in modifying asphalt binders to enhance its performances. The PPA is a liquid mineral polymer that can be utilized alone or combined with other additives in the bituminous mix. The utilization showed an enhanced rutting resistance and storage stability (Daly, 2017; Ameli et al., 2020). Daly (2017) specified that the PPA acts as a stiffening agent to remunerate the reduction in stiffness especially at high temperature ranges. Nuñez et al., (2014) and Behnood et al., (2017) has reported a better fatigue performance, stiffness, and shear modulus of both short-term and long-term aged asphalt binder with the addition of PPA. Furthermore, several researchers have reported the improvement made by incorporating gilsonite with the most optimum usage of 8% by weight of bitumen (Ameri et al., 2011; Jahanian et al., 2017). It is proven that gilsonite can improve the mixture performance in terms of Marshall Stability, resilient modulus, and rutting resistance according to Jahanian et al. (2017), as well as the high-temperature performance based on Ameri et al. (2011) research outcome.

2.1.2 Applications of Waste Materials & By-Products as Alternative Modifiers

Throughout the world, utilization of waste materials in asphalt technology has been acknowledged as a proficient method for years. The significant amount of solid waste generated from various sources such as household, agricultural, construction, industrial, municipal, mining, etc. have led to severe landfill issues. By referring to Global Waste Generation - Statistics & Facts (2020), the municipal solid waste (MSW) is foreseen to escalate 70% to 3.4 billion metric tons in 2050 due to urbanization, modernization, and population growth. Kaza et al. (2018) investigated that the regions of East Asia and Pacific, and the Europe and Central Asia generate the most waste, which consists of 43% of the world waste generated as shown in Figure 2.1 meanwhile the regions of Middle East and North Africa generates 129 million tonnes which is the least among the others as shown in Figure 2.2.

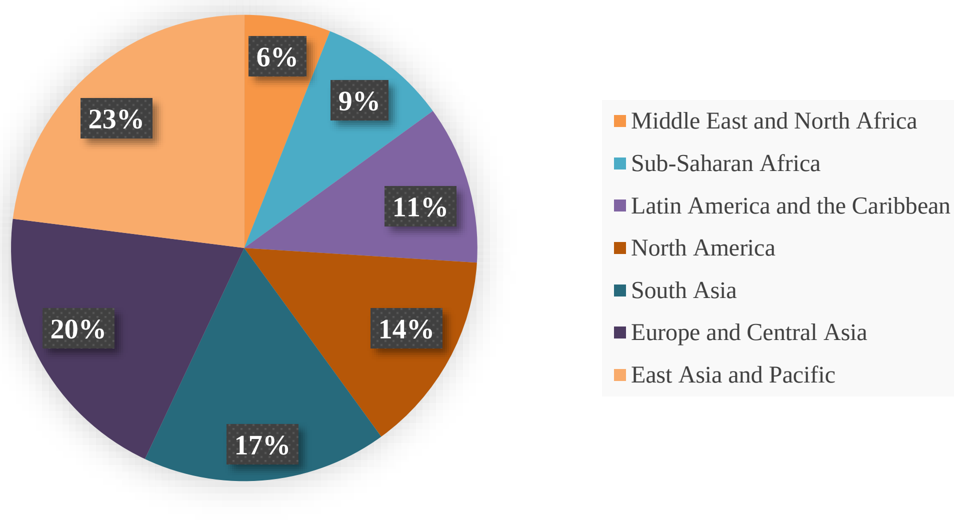

Figure 2.1: Percentages of Waste Generation by Region (Source: Kaza et al., 2018)

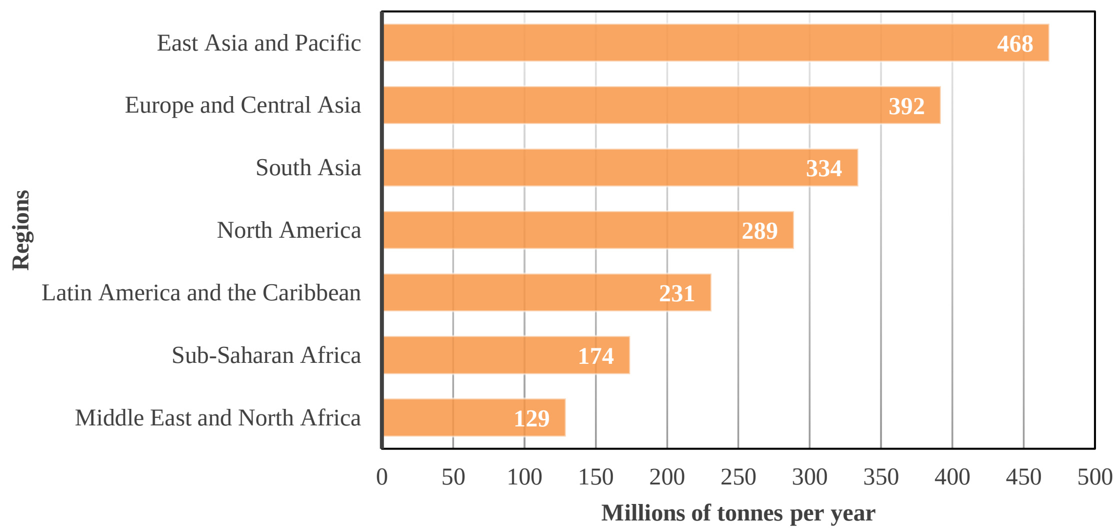

Figure 2.2: Waste Generation by Region in Figures (Source: Kaza et al., 2018)

Therefore, this urged the asphalt industrial players and researchers to implement the secondary or recycled, instead of primary materials into asphalt binder and mixtures to ensure a safe disposal and towards the sustainable construction.

Various recycled materials or by-products are adopted and implemented in pavement construction such as plastic waste, recycled polymer, rubber waste, steel slag, glass waste, fly ash, cement kiln dust, asphalt shingle, bio-oil, construction and demolition waste, rice husk and straw, paper waste, waste cooking oil, food waste, cigarette butts etc. (Antunes et al., 2017; Bamigboye et al., 2021; El-Badawy et al., 2019; Huang et al., 2007)

Waste materials such as plastic and polymer-based modifiers have been adopted as asphalt modifiers for a long period of time. The utilization is used to replace the virgin materials due to the high cost and recycled polymers are also found to exhibit comparable performances in modifying bitumen by Kalantar et al. (2012). The implementation of plastic in improving the asphalt mixtures has been demonstrated in Canada as an additive in warm mix asphalt (WMA) (Ridden, 2012), the Netherlands in road construction (Saini, 2015), and New Zealand by using plastic waste containers (Parkes, 2018).

In the past few decades, waste tire rubber has been implemented as additives via wet or dry process in asphalt mixtures. Nichols (2020) evaluated that recycled tire rubber is beneficial to asphalt binder modification as it promotes energy savings and emissions reduction. It is reported that China is the world's primary producer of waste tire rubber and it generated approximately a 14.58 million tons of waste tires in 2018 (Wang et al., 2020). Therefore, it has been developed into asphalt in Southern Beijing as the largest incorporation of recycled material in Asia (China Focus, 2019). Besides that, Tire Stewardship Australia has also promoted the use of recycled tire rubber in roads as mentioned by Nichols (2020).

According to World Steel Association (2018), 170kg of steel slag is produced per tons of crude steel and 630 million tons of scrap that produced from virgin steel are recycled annually. Steel slag has been utilized in concrete and asphalt as aggregate form in UK due to the high disposal amount produced from the steel production (Huang et al., 2007). In 2019, Department of Urban Planning and Municipalities (DPM) has implemented steel slag for roads paving in Abu

Dhabi. It has been utilized as aggregates form to modify the asphalt layers to obtain a better pavement performance and ensure infrastructure sustainability (Oommen, 2019).

2.2 Types of Waste Materials/By-Products Used for Asphalt Binders and Mixtures Modifications

2.2.1 Household Waste

Household waste or domestic waste generally consists of disposable materials that generated from households. This waste can be classified into two categories, i.e., non-hazardous waste (food waste, glass, plastics, etc.) and hazardous waste (batteries, household cleaners, toxic materials, etc.). Razali et al. (2020) stated that municipal solid waste is mainly sourced from waste generated from households in which 80% of the recyclable waste ends up in landfills. Consequently, it gives rise to environmental problems and leads to scarcity of landfill capacity. Asphalt pavement industries have explored various types of household waste materials to be recycled for the use in asphalt mixtures to minimize the environmental impacts.

Household wastes that commonly utilized as the asphalt binder modifier constitutes food waste, plastics, glass, electronic waste, etc. Generally, food waste can be yielded from organic sources such as fruits, vegetables, dairy products, and meat. Mahssin et al. (2021) has highlighted the application of liquefied bio-product which is produced via hydrothermal liquefaction of food waste such as cooked rice, vegetables, and chicken to be used as a potential non-petroleum-based binder in bituminous mixture. Eggshell powder (ESP) has also been utilized in natural asphalt (NA) along with the addition of commercially available LDPE according to Nejres et al. (2020). Besides, Jalkh et al. (2018) examined the recycling activity of spent coffee grounds oil (SCG) into

SCGs oil and used it as asphalt binder rejuvenators roadways with light traffic and for enhancing asphalt performance.

Plastic wastes such as polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET) and polyvinyl chloride (PVC) can be recycled and utilized to improve the performance of asphalt binders or mixtures in terms of aging resistance, resilient modulus, adhesiveness and reduce the cost of road construction (Baghaee et al., 2012; Singh et al., 2012; Rahman et al., 2013; Dalhat et al., 2016; Leng et al., 2018; Rahman et al., 2020).

Recycling of waste cooking oil (WCO) that origins from frying activity at high temperatures into bio-asphalt for the modification of binder is another sustainable way to reduce oil waste dumping issue (Wen et al., 2013; Azahar et al., 2016; Rahman et al., 2017; Geng et al., 2020). Moreover, numerous researchers (Zargar et al., 2012; Xingyu et al., 2020) have also evaluated the potential and performance of WCO as a rejuvenator for aged binders.

Glass waste which composes about 3% to 5% of all household waste has been recycled into glass-asphalt mixture which is also known as 'glasphalt'. Wu et al. (2004) recorded the earliest application of glasphalt is in late 1960s and it consists of a substitution of fine aggregate with crushed glass material in the asphalt mixture. In recent years, glass waste has been studied to incorporate it into bituminous mixture in various forms such as aggregates (Arabani, 2011; Shafabaksh et al., 2014), filler (Choudhary et al., 2020) and fiber (Mahrez and Karim, 2014; Baran and Karacasu, 2019).

Electronic waste or e-waste is generally yielded from discarded electronic devices and products. There are several researches conducted on the e-waste recycling and identification of its application within the asphalt industry. Typical recycled e-waste plastics includes Acrylonitrile Butadiene Styrene (ABS) and High Impact Polystyrene (HIPS) as asphalt binder modifiers

(Brennan et al., 2002; Hasan et al., 2016), waste printed circuit boards (Guo et al., 2009), waste polyethylene (WPE) film (Jeong et al., 2011), recycled computer plastics (Colbert and You, 2012), etc. The recycling activity is intended to ease the scarcity of landfill while developing a profitable application for e-waste plastics.

2.2.2 Agricultural Waste

Agricultural waste refers to the waste that generated from series of agricultural operations. It includes the crop residues that produced during the harvest and by-products that left unutilized after agricultural activities. (Nagendran, 2011; Choudhary et al., 2020; Fareed et al., 2020). This section discusses the modification of asphalt binder via the implementation of agricultural waste such as rice husk ash (RHA), palm oil fuel ash (POFA), sugarcane bagasse ash, groundnut shell, coconut waste, etc.

Rice husk ash (RHA) is produced as a by-product via the incineration of rice husk waste. RHA mineral filler in hot-mix asphalt (HMA) to replace the conventional mineral filler has been widely studied and applied in the construction industry (Sargin et al., 2013; Al-Hdabi, 2016). Besides, Mistry et al. (2019) examined the effect of using RHA to replace the conventionally used hydrated lime (HL) as fillers in hot mix asphalt (HMA). Fareed et al. (2020) investigated the performance of asphalt binder and mixture incorporated with nano agriculture waste such as RHA which undergoes pre-reduction of size to Nanoscale using a ball mill.

Every year, sugarcane production especially in Brazil has generated enormous waste known as Sugarcane Bagasse. The burning of the waste results in the production of Sugarcane Bagasse Ash (SCBA) which can be utilised as asphalt modifier to reduce the quantity of waste disposal. In 2018, Mansor et al. examined the performance of stone matrix asphalt (SMA) mixtures that incorporated with 0.3% of sugarcane bagasse fibres by total weight of mixtures. In the same year, Singh et al. has also developed a research using 10% and 15% Bagasse Ash (BGA) that is blended into reclaimed asphalt pavement (RAP) aggregates as partial cement replacement (Singh et al., 2018). Similarly, Ribeiro et al. (2014) evaluated the by-product of sugarcane ethanol industry or SCBA as a partial replacement material.

Malaysia as the main crude palm oil supplier in the world generated about 19.15 million tonnes in 2020 according to Malaysian Palm Oil Board (MPOB) (2020). The combustion of oil palm waste results in the generation of Palm Oil Fuel Ash (POFA). Rahman et al. (2017) indicated that the use of POFA as additive to modify asphalt binder has significantly enhanced the stability, flow and rutting resistance. Borhan et al. (2010) and Ahmad et al. (2012) examined the practical use of POFA as mineral filler at various percentages. Hainin et al. (2014) also studied the performance of asphalt pavement with the incorporation of POFA as bitumen modifier which is under regular conditions and subjected to aging.

Large quantities of groundnut shell ash (GSA) are produced from the combustion of groundnut shell which has 47.32 million tons worldwid0e annually as reported in United States Department of Agriculture (2021). GSA can be recycled and used in constructions to reduce the quantity of disposal and as an alternative source of replacement construction materials. Several researchers such as Arabani et al. (2020) assessed the suitability of applying GSA at different concentrations as an asphalt binder modifier meanwhile Edeh et al. (2013) conducted a research to characterize the incorporation of GSA to stabilize reclaimed asphalt pavement (RAP) for the use of flexible pavement materials.

Coconut is another common solid waste by-product that has been widely studied and implemented in asphalt mixture. Coconut has been utilized in different forms such as ripe coconut fibre in stone matrix asphalt (SMA) mixes (Panda et al., 2013), coconut shell in asphalt mixture which undergoes pre-treatment to reduce its hydrophilic properties (Ting et al., 2016), modified bitumen with nano-charcoal ashes generated from coconut shell (Jeffry et al., 2016) and coir fibre/coconut fibre in HMA mixtures (Khasawneh and Alyaseen, 2020).

2.2.3 Industrial Waste

Various industrial or manufacturing sectors produce waste from a diverse range of different processes such as ashes, slags, kiln dust, sludges, etc. This section highlighted the major waste generated from industries and their alternative application within asphalt mixture modification after recycled.

According to Muniandy et al. (2009), there is approximately 30% of the daily ceramic production contributes to the generation of ceramic waste (CW). Recycling ceramic waste as aggregates and mineral filler could be an interesting alternative instead of disposing it to landfills. In previous studies, Huang et al. (2009) and Binici (2007) used CW as fine aggregates. Feng et al. (2013) examined the effect of asphalt mixture that is added with different percentages of crushed ceramic waste aggregates (CWAs). From the studies, the thermal conductivity of asphalt mixtures has been reduced. Shamsaei et al. (2020) conducted a study to replace the conventional limestone filler with CW powder at various proportions. The result shows an improvement in Marshall stability, moisture resistance, fatigue life, rutting resistance, and mechanical properties of HMA.

Coal Fly Ash (CFA) is typically a by-product that produced by coal-powered plants for electric generation. From previous studies, the substitution of CFA as filler is proved to improve the properties of HMA and WMA such as ravelling, stripping resistance, aging resistance, workability, fatigue life, etc. (Sharma et al., 2010; Xiao et al., 2012; Mistry et al., 2019; Choudhary et al., 2020). Muniandy et al. (2009) have also investigated the suitability of CFA as mineral fillers in SMA mixtures and it showed a satisfied physical and chemical properties by achieving the quality requirements.

During the steel-making process, industrial waste such as steel slag (SS) is generated. It has been widely utilised in asphalt mixtures in aggregates form as mentioned by previous researchers such as Zhou et al. (2020) with the utilisation of SS as fine aggregates and Chen et al. (2020) as coarse aggregates. The studies indicated that SS coarse aggregates are effective in enhancing moisture stability, fatigue resistance, deformation resistance and skidding resistance. There are several studies (Li et al., 2016; Tao et al., 2019) have demonstrated the characterization of SS as mineral filler and it enhanced the deformation resistance of the binder.

Various applications of bauxite residues or 'red mud' as a substitution of alternative mineral filler in pavement structures have been investigated in recent years (Zhang et al., 2018; Choudhary et al., 2019; Zhang et al., 2020). Bauxite residues or known as 'red mud' waste is generally a solid by-product that generated during the refining of alumina. Zhang et al. (2021) highlighted that the incorporation of red mud is found to enhance the asphalt mastics and mixtures in terms of stiffness, mechanical behaviors and rutting resistance and being susceptible to moisture and ravelling action.

Nowadays, crumb rubber modified binder (CRMB) has been widely adopted and used as an alternative in enhancing hot mix asphalt performance properties and to overcome the disposal problem of waste tire rubber. Moreover, the aging, fatigue, cracking, skid and rutting resistance are being improved by implementing crumb rubber into the asphalt (Mashaan et al., 2014; Leng et al., 2018). However, the large quantity of crumb rubber powder possessed high viscosity, resulting in limited application (Yu et al., 2017). Therefore, a proper dosage of CR is required to satisfy both pavement performance and construction standards (Cai et al., 2019).

2.3 Development and Performance of Bituminous Materials Modified Using Various Recycled Materials Worldwide

Throughout the world, there is always an issue regarding landfills and natural resources exploitation, which causes the scarcity of virgin materials, high transportation fees and excessive aggregate mining. Previous years, asphalt pavement industries and government has been exploring the opportunities and benefits of waste materials implementation such as construction and demolition (C&D) aggregates, RAP, steel slags, fly ashes, and municipal solid waste products in asphalt pavements (Schimmoller et al., 2000). A sustainable pavement has shown a minimized exploitation of natural resources, lowered energy demand, limited greenhouse gas emissions and improved safety and health. Hence, new alternatives to promote the use of recycled waste materials in substituting the use virgin materials are investigated and developed in most countries.

In this section, development, and performance of the incorporation of waste materials in bituminous materials in countries such as Asia, North America, South America, Europe and Africa has been reviewed.

2.3.1 Asia

According to Kokotovic (2021), India has included waste material such as plastic in road pavement construction in the past two decades ago. The 60,000 miles of plastic roads have been found to have higher tolerance towards heavy loading, rutting, moisture sensitivity, cracking and temperature susceptibility.

Besides, an average of 73% RAP has been utilized in asphalt mixtures along the southwestern side of Tokyo Bay. The incorporation of RAP content has constantly increased from 32.5% (2000) to 47% (2013). Asphalt recycling aimed to minimize waste generated from asphalt paving work, preserve natural resources, conserve energy for transportation and extraction (West and Copeland, 2015).

In Malaysia, Public Works Department's Centre of Excellence for Engineering and Technology (CREATE) is recently monitoring on a project of road pavement technology that implemented plastic waste and latex in asphalt at Kerdau, Temerloh, Pahang; and Chukai, Kemaman, Terengganu. Plastic waste is generated in factories and shopping malls and is processed before being used (Kamarudin, 2019).

Iron and steel slag have been implemented in base course material and the mixture is appointed as a procurement item by Green Procurement Law. The incorporation of the iron and steel slag has been adopted in the asphalt construction work in Japan such as Kobe City, Himeji Higashi ramp on Sanyo Expressway and Highway 170 at Osaka Loop Line. It is found to be effective in preserving environment and enhancing the stiffness as well as wearing resistance of the asphalt mixture (Nippon Slag Association, 2014).

2.3.2 North America

Recent years, California has developed the implementation of crumb rubber for asphalt pavement use. California Department of Transportation (Caltrans) has utilized 35% of tire rubber for being recycled and used in their paving projects. The binder that used is required to be replaced by 18-20% of rubber by weight. As a result, the rutting, cracking and aging resistance have been enhanced by the modification of tire rubber (McElvery, 2021).

Besides that, TechniSoil in Los Angeles has replaced the bitumen with plastic waste. It is reported that 2,300kg of recycled PET plastic has been implemented in 1.5km two-way road as to conserve carbon resources and enhance the asphalt durability (Ralston, 2020). In United States,