AN OVERVIEW ON NATURAL RUBBER APPLICATION FOR ASPHALT MODIFICATION

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ABSTRACT

Natural rubber with asphaltic materials is a great invention towards producing an admirable quality of asphalt paving roads. With its inherent chemical composition as elastomer kind of properties, this polymer modification can change the performance of current conventional asphalt paving roads which is recently faced limitation against temperature changes and traffic loading. Consequences, such as global market price of rubber turns in very uncertainty ways, has encouraged the government to seek for an alternative utilization of the natural rubber to backing up the industry. Therefore, natural rubber is seen as a preference material to multiply the quality of asphalt for road construction as well as to support the economic growth of the rubber industry. This paper provides an overview of natural rubber that has been applied as an additive in asphalt binder transformation. Numerous studies have been carried out on natural rubber based on different aspects of compatibility and interactions between the polymers and the asphalt aiming to improve the performances of the modified materials. From the review, the natural rubber used in a form of latex concentrated, Ribbed Smoked Sheet (RSS) and Liquid Natural Rubber (LNR) show promise in improving the asphalt mixes. Therefore, further study is needed to make them applicable as rubberized asphalt binder, thus provide better performance and services on the asphalt pavement.

Key words: Asphalt; Latex Concentrated; Ribbed Smoked Sheet (RSS); Natural Rubber; Alternative Utilization

Introduction

Natural rubber technologies are becoming wider spread in numerous applications in enhancing its usage on various forms of products. Malaysia is a leading producer country of rubber after Thailand and Indonesia. Currently, rubber is harvested mainly in the form of latex tapped from rubber tree. In addition to its raw rubber compound, water and minor impurities such as proteins and resins are included in this latex form (Hamdan *et al.,* 2000). This engineering material has been successfully attempted for many years in the production of tires, gloves (Suhawati *et al.,* 2014), as well as material for building and road constructions. As

road material, with its organic polymer compound behavior, natural rubber becomes one of the alternative solutions in improving the asphalt properties for asphalt pavement. Asphalt, on the other hand, is a natural substance derived from the distillation of crude oil (Arafat and Rosli, 2011) and has a viscoelastic property which is quiet sensitive to temperatures fluctuation (Saad *et al.,* 2013). This simply means that conventional asphalt, without any modification, it tends to have permanent deformation and at the same time reduced pavement service life and increase expenditures in maintaining road performance (Arafat and Rosli, 2011). Recently, due to global commodities slump, rubber price falls even more. The price of natural rubber was fluctuated and the production was overcapacity and imbalanced which therefore need for variation in its application to help the rubber industry (Nopparat *et al.,* 2012). Among the natural rubber used in asphalt modification from previous research are mostly in the form of latex because mixing with natural rubber latex provides the most efficient product compared with other form in equal quantity (Smith, 1960) and some other used Ribbed Smoked Sheet (RSS) and Liquid Natural Rubber (LNR). In spite of its effectiveness as a modifier, this potential application seems to increase domestic consumption as much as possible in order to add value to domestic natural rubber dealing and at the same time give better overall performance in the bituminous mixes toward better flexible roads (Nrachai *et al.,* 2005). Therefore, this paper gives an introduction of natural rubber, manufacturing process and its potential application as an additive for asphalt modification. It is an attempt to provide sufficient information on the aforementioned topics in engaging over researchers and industrial interests.

Natural rubber

In early development of natural rubber, it is developed in latex form from rubber trees, which is, by provenance, it is an agricultural commodity, mostly belonging by the spurge family name *Euphorbiaceae* (Sethuraj and Ninan, 2012). Natural rubber occurs in almost 2000 plant species, only a few of which are industrial important, the first place being occupied from a tree which is autochthonous to the tropical rain forest name as *Hevea brasiliensis* (Ikeda and Kohjiya, 2014; Franta, 2012). This species is broadly used due to the fact that it is grows well under cultivation and with proper management of tree wounding, more latex can be produced. Other plants that also been used for commercially exploited natural rubber are Panama rubber tree (*Castilla elastica*), rubber fig (*Ficus elastica*), vine rubber (*Landlphia*), gutta-percha (*Palaquium gutta*) and *Funtumia* (Ikeda and Kohjiya, 2014). There are some rubber-bearing species which is classified as source of rubber such as *Taraxacum* species including *Taraxacum officinale* and *Taraxacum kok-saghyz*; cultivated by Russian scientist, and guayule (*Parthenium argentatum*); cultivated by the American scientist due to its hypoallergenic properties (Ikeda and Kohjiya, 2014; Sethuraj and Ninan, 2012). These rubber sources required extra processing and some are difficult to tap make them not much effective in rubber industry.

Natural rubber exhibits unique and special chemical and physical properties and it is greatly influenced by the presence of non-rubber substances (Thomas *et al.,* 2013). Table 1 shows the composition of the Natural Rubber. In view of its chemical properties, which normally in latex appearance, this polymer make up in cis-1,4-polyisoprene with a chemical formula of C_5H_8 that composed of carbon and hydrogen atoms alone with high molecular weight (Thomas *et al.,* 2013; Ikeda and Kohjiya, 2014). Due to its double bond presence in each repeated unit, polyisoprene can be created synthetically (synthetic natural rubber) and also susceptible to undergo vulcanization. This is a process of adding sulphur into latex to improve raw rubber constituents in which change the thermoplastic elastomer properties of natural rubber into a thermoset (Krevelen and Te, 2009). In depth reviews of its physical properties, natural rubber largely exhibits elasticity due to its high stretch ratio and resilience characteristic. This happens due to a lot of complex molecular chains of polyisoprene, when under loading, performed almost linear chains and wrinkled back to its position when the load is removed (Mark, 1996). Rather than that, viscoelastic properties of rubber are also discussed due to the fact that it combines both liquid-like and solid-like characteristics (Payne, 1966). As it is agricultural product, natural rubber is a renewable material and the only biomass that strain crystallizes that occur spontaneously at low temperature or when it is stretched, hence lowering the elasticity value (Ikeda and Kohjiya, 2014). Since vulcanization is suggested, rubber creates disulfide bonds between chains, limiting its degree of freedom and the chains speedily tighten. Thus, the rubber is harder and less extensible because of increasing in elastic force (Studebaker, 1966). It should be noted that the final properties of rubber item depends highly on modifiers and fillers such as carbon black, factice, whiting and a host of others instead of the polymer itself.

Table 1: Composition of Natural Rubber (Bhowmick and Stephens, 2000)

Component	Percentage by weight
Rubber Hydrocarbon	93.7
Lipids	3.4
Proteins	2.2
Carbohydrates	0.4
Ash	0.2
Others	0.1

Processing of natural rubber has slightly expands the application on other raw rubber products. After tapping the rubber tree process, latex will be collected into cups to prevent from flowing into other part of the tree. There are two main methods involving rubber field latex; the first one undergo ammoniation (adding high amount of ammonia concentration) for latex preservation before patterned in soft solid slabs rubber, while others by kept the latex in cups for certain period of time until it

coagulates by itself or adding formic or acetic acid to faster turn to "cup lump" (Groover, 2007). Coagulated latex can then be processed into higher-grade forms such as block rubber, ribbed smoked sheet, pale creep and air-dried sheet (Bhowmick and Stephens, 2000) while naturally coagulated cup lump is used in the manufacture of TSR10 and TSR20 grade rubbers (Franta, 2012). In recent years, Asia is the main source of natural rubber and Thailand, Indonesia and Malaysia are the three largest main producing countries natural rubber production. There are about more than million tons of rubber produced every year, which by approximation, up to 40 % was in natural state and the rest used for synthetic variety rubber products (Jacobson, 1983). Present day science and technology has led to new liquid natural rubber, sheet creep types of natural rubber or other modified forms into more commercial aspects to see the potential instead of using raw rubber only.

Manufacturing And Application

Rubber manufacturing use huge amount of natural rubber to produce high quality rubber end products. Compounding rubber with additives is the unbeatable method designed to satisfy the given application in terms of properties, cost and process involved and was formulated for rubber to produce end products like tyres, belting, engine and other mechanical goods (Sethuraj and Ninan, 2012). Vulcanization is a must process that generally applied to rubber or elastomeric materials to achieve rubber compounding by reaction with sulphur and accelerators at higher temperature. Vulcanization converts rubber from plastic-temperature sensitive materials to technologically-useful elastic materials by the formation of a cross-linked molecular network (Sethuraj and Ninan, 2012; Mark, 1996; Roberts, 1988). Other than vulcanization, compounding of rubber by incorporation with additives, fillers and small amount of oils helps to protect rubber compound against ozone, heat and oxygen as well as to increase modulus, strength and reduce the processing cost (Sethuraj and Ninan, 2012).

Vulcanized rubber had high ranking demand since the Second World War and hence turns down its supply number. Simultaneously, the synthetic polymer industry came out as a main market force and rising up rubber commercial value (Sethuraj and Ninan, 2012). Synthetic rubber, which is made by the polymerization of a variety of petroleum-based precursors, was used largely in the manufacture of automobile tyres, conveyor belts, noise and vibration insulators for marine products and miscellaneous rubber goods (Woodard, 2001). Examples of synthetic rubber produced are including silicone rubber, butyl rubber, nitrile rubber, chloroprene rubber, foam rubber and the most usually operated is styrene butadiene rubber (SBR) (Woodard, 2001). In Malaysia, synthetic rubber company has introduced polybutadiene rubber (BR) chemical production as the general purpose is to expand the usage on tyres and industrial rubber products (Goldthorpe, 2015). The consumption by production sector is given in Table 2.

Table 2: Consumption by production sector: 2012 (Goldthorpe, 2015)

Tyres	**General products**	**Latex products**	**Total**
Metric tons	Metric tons	Metric tons	Metric tons
56,514	80,189	562,367	699,070

Note: General rubber products include rubber goods, general rubber goods and footwear.
Source: MRB 2013

Another chemical technique to develop rubber applications is by modifying natural rubber straight after it is harvested from the tree. This is because the natural rubber has a fixed structure of cis-polyisoprene and cannot be polymerized by itself to provide preferable pendent groups like synthetic rubber (Bhowmick and Stephens, 2000). Many modified rubber have been prepared with variety of functional groups, where each of them has its own special cross-section (Klempner, 1994). The modified physique will include hydrogenated NR, chlorinated NR, hydrohalogenated NR, cyclized NR, epoxidized NR, resin-modified NR and etc. (Bhowmick and Stephens, 2000; Kalia and Avérous, 2011). Some of these modifications were then be replaced by other materials, thus the production has decreased into small amounts to prevent wastage (Rowe, 1989).

Asphalt modification

The performance of asphalt pavement is mainly governed by the properties of the binder. In view of this factor, discovering on pavement constituents like asphalt and its commodity are much preferred. Asphalt is one of the oldest known engineering materials (Whiteoak, 1995). Other than used as adhesive, asphalt is also used in various ways as sealant, preservative, waterproofing agent and pavement binder (Jiqing *et al.,* 2014). The use of asphalt has increased rapidly in world consumption, where most of which was used in road construction. In addition, the asphalt properties are closely related to the crude oil sources and depend on refinery process. This then contributes asphalt chemical composition which is very complex and variable. Thus, by selecting good crude oil sources or having excellent refinery process, it can produce good asphalt quality at the same time. However, the limited oil resources and low effective control during distillation failed to produce extraordinary asphalt, hence, influenced other factors to improve high earnings in asphalt industry through asphalt modification (Becker *et al.*, 2001). In fact, pavement fields have been developed faster all around the globe during the last few decades, substantially in developing countries.

Following the rapid development, road pavement face challenges due to increasing traffic load, higher traffic volume and insufficient maintenance led to severe failure on the surfaces and reduce the service life. Asphalt behaviors are closely related with temperature and loading time (Arafat and Rosli, 2011). In tropical countries, asphalt is soft at normal temperature but tends

to flow like viscous liquid at high temperature. Here, the lack of rheological and viscoelastic properties of asphalt can cause deformation on road surface i.e. rutting damage that take place along the wheel part (Nopparat *et al.,* 2012; Vasavi and Durga, 2014; Billiter et al., 1997). Otherwise, at low temperature, asphalt change it properties to become hard and rigid solid cause brittleness and led to fatigue cracking failure (Al-ani, 2009; Peralta *et al.,* 2014). This is also supported by other studies, which mentioned that the conventional asphalt pavements tend to experience a lot of pavement distress especially fatigue failure, rutting as well as moisture damage (Chen and Huang, 2008; Lundstrom *et al.,* 2004). Thus, study conducted by Sengoz on effect of asphalt film thickness on moisture sensitivity cleared that with the increasing of asphalt film, effect of water can be deduct and strength of asphaltic mixture is higher (Sengoz and Agar, 2007). These problems made highway engineers realized about the methodology in finding the solution to overcome pavement matters.

Asphalt modification is one of the approaches that can be considered to improve the performance of asphaltic pavement. Different modifications to asphalt are being explored such as modification with Sasobit wax, carbon black, mineral fiber and others. In order to get a bituminous conglomerate with premier renditions asphaltic pavement, Polymer Modified Asphalt or PMA is one of such improvement by mixing polymers in asphalt to produce a superior form of road pavement (Al-ani, 2009). The advantages of PMA include, increasing in viscosity and elasticity (Al-Mansob *et al.,* 2014; Nrachai *et al.,* 2005) of asphalt with higher softening point and aging resistance as well as to reduce distresses on asphalt pavements (Peralta, 2012). Among the polymers used, natural rubber is an interested polymer that can be efficiently added into asphalt (Nopparat *et al.,* 2012). The distingue quality of natural rubber is its high stability, excellent tear strength and fatigue resistance, extending of the durability of asphalt pavements (Tayfur *et al.,* 2007). In addition, natural rubber has excellent dynamic properties with a low hysteresis loss, good low temperature properties and has strong bonding to metal parts (Vasavi and Durga, 2014). Therefore, the improvements made on the quality of asphalt for road construction is practical to prolong lifetime of roads and save the maintenance cost (Polacco *et al.,* 2005).

Natural rubber – asphalt blends

The concept of using natural rubber latex has been applied in asphalt surfacing for over 30 years and is perceived to improve asphalt performance. Additionally, adding natural rubber latex is such a good method due to their characteristics that contain discrete rubber particles which available to fused with asphalt easily (Robinson, 2005; Franchen, 2004). During cold weather, freezing asphalt can cause crack under stress. Thus, by applying latex into pavement modification, this raw rubber acts like an elastic band that helps dissipate developed stresses and grip asphalt altogether at the same time. As the temperature increases, asphalt rheological properties change drastically hence it begins to flow. Herein, the function of latex as polymer acts like membrane, resists the asphalt flow and enhance the shear resistance (Wen *et al.,* 2015). Its inherent elastomer properties proved that natural rubber latex has high potential in improving long-term pavement performance through asphalt concrete mix (Shaffie *et al.,* 2015).

Latex, or normally known as field latex, contains about 36% rubber including small percentage of impurities and the rest is water. However, in some cases, field latex could only maintain its liquidity maximum up to three hours long (Nrachai *et al.,* 2005). In view of this factor, preservation of field latex is needed for long term storage, as it is an important raw material with a wide range of applications. Recently, many researchers have been conducted asphalt binder modification to improve asphalt properties by using preserved latex as modifier. In a further study by Fernando, M.J. Nadarajah, M (1969) had analyzed on improvement asphalt properties by using miscellaneous raw latex rubber such as field latex, concentrated latex and skim latex. Results founded were related with increment of softening point and penetration value reduction. Next, a study conducted by Nrachai *et al.,* (2005) showed that natural rubber latex is the best alternative for pavement making contributes to the elongation of service life expectancy due to improvement on its flexibility and stability properties. Thus, it demonstrates that the roads layered with rubber-modified asphalt shall have more strength and resistance than the conventional asphalt. Furthermore, Krishnapriya found that performance of natural rubber modified asphalt mixture gives better results compared with the ordinary asphalt mix due to result of rubberized asphalt whereas not only increases rut resistance, but also ameliorate fatigue life with the increase in resilient modulus (Krishnapriya, 2015). Research conducted by Shaffie *et al.,* (2015) on evaluating Hot Mix Asphalt (HMA) stripping performance using natural rubber reveals on the improvement of physical properties and pavement performance thus, concluded that natural rubber is a good modifier to enhance the asphalt binder characteristics.

Research performed by Nrachai *et al.,* (2005) on modification of asphalt with natural rubber latex used both Marshall Method and Gyratory Compacter to determine optimum binder content at two distingue percent of air void. The results showed that the natural rubber could increase the density and stability of asphalt mixture and less optimum binder content is required. On performance properties, adding latex can reduce stripping of aggregates and increase its resilient modulus thus enhance fatigue and permanent deformation. In another study by Saad *et al.,* (2013) on combination of natural rubber and carbon black found that the modification has improved paving mixture stiffness and enhances its rutting resistance. Study conducted by Krishnapriya (2015) on performance evaluation of natural rubber also proved the rutting resistance and fatigue life of road pavement had increased. Furthermore, Shafffie *et al.,* (2015) studied stripping performance and volumetric properties of asphalt mixture using natural rubber and granite aggregates found that local aggregates meet both Superpave consensus aggregate and sources aggregate requirement. This shows that natural rubber polymer that was added to the binder coat aggregates very well thus improving stripping resistance on pavement surface.

Apart from the utilization of natural rubber latex in asphalt modification, rubber sheet is also one of the applicable materials to mix with asphalt. Despite of its different shape and form, rubber sheet has its own method in adding into asphalt content. In consideration of its chemical constituent, rubber sheet has similar properties with natural rubber latex and it is suitable to combine with asphalt due to less water content and as additive in road application. Examples of rubber sheet products are Ribbed

Smoked Sheet (RSS), creep rubber and block rubber (STR 20) (Jawjit *et al.,* 2010). This conventional form of natural rubber can influence road performance such as increasing in viscosity, make road pavement more strength and enhance its durability (Al-Khateeb and Ramadan, 2015). In addition, adding rubber sheet can prevent frothing and foaming to occur while blending it with asphalt. Study conducted by Nopparat *et al.,* (2012) discussed that by using Ribbed Smoked Sheet as natural rubber sources greatly influenced its binder properties. It was found that the roads paving with natural rubber-modified shall have more durability and strength than using conventional asphalt. Furthermore, torsional recovery test has enhanced asphalt performance in elasticity thus affect the toughness and tenacity properties at the same time. However, the ratio of the RSS added must take into account since over-content of rubber can cause mixture less homogeneity thus affect the binder properties.

Instead of using latex, Liquid Natural Rubber (LNR) is such an improvement in rubber properties as LNR can provide more homogeneous binder, make it easy to blend with asphalt as foaming and frothing problems due to water evaporation can be distracted with the elimination on temperature falls (Okieimen and Akinlabi, 2005). Availability of natural rubber in this form will increase its competitiveness in relation to synthetic rubbers, thereby enhancing benefits and value of rubber application. The LNR was previously found to have the most of neutral components as its production reduced the molecule size of natural rubber by depolymerization of latex using nitrobenzene or via oxidation of H_2O_2 and $NaNO_2$ as reagents (Boochathum, 1985). Apart from that, LNR can be produced by using peptizing agent before being heated in a depolymerizer. Nair *et al.,* (1992) had studied on improvement asphalt properties using fumigated rubber to reduce molecule by dispersing sheet rubber into fluxing oil after mastication process to convert into LNR and then been mixed with asphalt cement by heating. It was observed that adding LNR reduced ductility and enhanced softening point (Tinavallie, 2013). The ratio of mixing both materials must be adequate and balance to prevent less homogeneity and over viscous at the end of the process. Results obtained from performance and binder properties tests show that the physical strength of natural rubber is decreased due to depolymerization process and adding LNR exhibits more viscous properties rather than elastic.

Challenges And Recommendations

In expanding natural rubber applications on various products and usage, obstacles to go through the process of successful must be eliminated in order to obtain good results. The global rubber industry has always been concerned over some factors, which can be described as challenges, in facing natural rubber development. Thus, the major barriers that influenced the growth on this material towards better establishment are subjected on natural rubber volatility price, competition from palm oil and higher request for synthetic rubber.

Price is certainly the first factor in the eye of consumers that had to take into account in order to gain more profit and at the same time producing high quality products. Although the industry may not be in shortage of primary source of natural rubber, yet the prices have been very uncertainty, contributing to rising prices for tyres and other end products of rubber. The reasons are due to unsynchronized natural rubber demand to its production (Nrachai *et al.,* 2005). In fact, natural rubber output was overcapacity during economic crisis, make them not properly visible in rubber market at higher rates and affects negatively on industry competitiveness. Palm oil plantation is also one of the raw materials industry that growing up simultaneously with natural rubber in some of the Southeast Asian countries. Thus, this eventually creates competition among both of the materials and will affect each other supplies on world demand. Since two decades ago, palm oil emerged as the cheapest source of edible oil and has been garnering a lot of attention on industrial global trading.

Natural rubber is a highly valuable biomaterial in contrast with other biopolymers and because of their special pure characteristic, natural rubber has been commercialized into synthetic rubber. This is happens due to the supply of natural rubber that has never satisfied demand for rubber and the gap had to be filled by synthetic rubber. It may be noted that the end products of synthetic rubber like heavy duty truck and aircraft tyres, gain higher world demand rather than natural rubber due to its physical and chemical properties that had change during the production. In order to defeat those problems on natural rubber challenges, actions need to be taken in a short time before this raw material went lost on its economic value.

Currently, another form of raw rubber i.e. cup lump rubber has been also identified to be used as an additive to the conventional road materials. Cup lump is a freshly coagulated rubber where the coagulation process takes place in the cup at the tree (Nrachai *et al.,* 2005; Fernando and Nadarajah, 1969) is seen as a new alternative on long-term solution to increase domestic consumption of natural rubber through its application with asphalt materials (Zborowski and Kaloush, 2007). Hence, at the same time, it can increase natural rubber usage over synthetic rubber and could also create healthy competition with other raw material sources. In view to the aforementioned advantages, previous research encountered various challenges on finding the suitability in mixing natural rubber and asphalt for modification of road. It should be noted that blending interaction gives high impact on natural rubber physical properties and also on bituminous mixture (Kintali and Anirudh, 2015). The biggest challenge that prevents more extensive use of natural rubber used in road construction is the lack of performance tests that accurately demonstrate the affects that polymer modification has on asphalt. Therefore, it is reasonable to study more on suitable kind of rubber used for different modifications, the ratio of natural rubber mixed with asphalt and techniques applied i.e. various temperature used and the mixing time in producing rubberized asphalt mix (Ibeagi *et al.,* 2012; Selvavati *et al.,* 2002).

Conclusion

To overcome conventional asphalt mixture problems, modifications with additives could become an alternative way to save asphalt source. In this respect, there have been some recent natural rubber attempts to expand it applications into more productive way against the limitation on its content through combination with bitumen. This review highlights past research that have been conducted on it advances and encountered challenges in the field of bitumen polymer modification that allow natural rubber to

influence binder properties, thus establish high quality of asphalt mixture. From this polymer-asphalt modification, previous research has approved that rubber is one of the materials that can be used as paving material to overcome road problems such as aging, rutting, fatigue failure to facing challenges due to current temperature and load from vehicles. Parallel to the uncertainty in global market price of rubber, addition of this raw material can help in support the economic growth of the rubber industry since the cost is lower than other materials and in fact, such application could potentially improve the quality of road, extend its service life, and reduce expenditures in maintaining road.

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