Recycling of waste rubber as fillers: A review

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Abstract. Recently, in line with raising environmental concerns, researchers are now focusing on the uses of recycled product. Waste rubber is one of the waste material that has been recycled and being used in many area such in civil engineering, tyres production, polymer composite, energy source and many more. This paper highlighted the concern on the raising numbers of waste rubber products generated globally and how does this waste rubber can cause a serious threat to the environment. Re-use, recycle, recovery and pyrolysis is the common method in ensuring sustainable management of waste products and it keeps emerging. This paper will discussed briefly the development of recycling waste rubber into a composite material, the methods, characterisations and improvements that contributed by the waste rubber on the composites properties. Focus is on the development of composite filled with waste rubber tyres, waste rubber gloves, waste rubber condoms and few other waste rubber products. Incorporation of waste rubber fillers has improved the mechanical and physical properties of the composites. Thus, composites with waste rubber fillers has created a new sustainable material and has potential to be applied into various field.

1. Introduction

The constant increase of rubber waste has become major environmental problem for countries around the world. This is due to the demand of rubber products keep increasing, as for example the outgrow numbers of vehicle production and also high demand of rubber usage in industrial, medical, engineering, household products and many more. Due to that rising numbers of discarded rubber products, disposing these substances becoming a burden to the municipal council and lack of environmental awareness among citizens makes the problems of disposing rubber products worsen. Land fill dumping and open burning are among common, cheapest and easiest method of disposing waste rubber which leads to water, air and soil pollution. The poor disposal of rubber products globally becoming a potential threat to human health or even the living being and decrease the quality of environment [1-2]. Shu and Huang [3] reported that due to large volumes of rubber scraps,
Landfilling was no longer able to keep up with the increment as land cost are hiking up, thus disposing waste rubbers becoming a highly costly method.

Like other polymer materials that provides tremendous benefit to humankind, rubber is one of essential materials in many application due to its unique and impressive properties. Rubber exhibit outstanding behavior that is very high elasticity properties as it can be stretched to a few hundred percent and recover almost instantly to its original shape when force being released [4-5]. Extremely high elasticity behavior of rubber provides by remarkably low Young’s modulus and high yield strain which resulted from the vulcanization process [6]. Rubber having thermostet cross-linked structure which provides a very durable properties and high resistance to environmental agents. Due to these outstanding properties, rubber are widely used in application ranging from household, healthcare, industrial, military, automotive, civil and outer space application. However, these outstanding properties has a setback that causing rubber very difficult to degrade due to its complex structure and composition.

Rubber also held another valuable characteristics such as able to dampen or reduce vibration [4-5], better wet grip, relatively resistance to gas and water permeability and reduced swelling in hydrocarbon oils [7-8]. Another essential properties of rubber are high resistance to acids, alkali and chemical solution. It makes rubber an important materials in many critical environment including healthcare, food processing, chemical manufacturing, refining, automotive, electronics and semiconductor manufacturing, pharmaceutical and research laboratory [7-8].

The above mentioned properties of rubbers making rubber among the high demand materials as reported by the Malaysian Rubber Export Promotion Council (MREPC), the total world production of rubber in 2016 has increased to 26.9 million tonnes million tonnes if compare from 2010 [9]. International Rubber Study Group (IRSG) also forecasted that the world total rubber consumption will increase annually at an average 2.8% per annum from 2017 - 2025. In 2017, IRSG forecasted the world total rubber consumption is 28.05 million tonnes [10]. These numbers may raise concern among environmentalist as it indicates more rubber waste will be generated.

However, many literatures shows that a great progress has been made in sustainable management of waste rubber. An important effort taken by the researchers to ensure sustainable management of these rubber wastes are through reuse, recycling, recovery, and pyrolysis. Reuse the worn rubber (i.e: tyres) simply by retreading and reuse back. Recycling of waste rubber includes utilizing the discarded rubber into various applications such as in marine conservation program (as an artificial reef, erosion control, breakwaters and floatation devices). Other than that, recycling method includes grinding the waste rubber and then incorporate the rubber particles into matrices such as concrete or polymer and used for civil engineering, or being manufactured into brand new products such as athletic tracks and playground surface, as filler for rubberized composite and many more [5, 11]. Besides that, recovery is a method where the waste rubber is used as a fuel source for high temperature process such as cement kilns, steam production, electrical energy and etc. While, pyrolysis is to yield the basic chemicals in rubber waste by decomposing the rubber component such as steels, gas, oil and its filler such as carbon black [11–13]. Table 1 shows that some of the applications for waste rubber in our daily life.

### 1.1 Waste rubber types

Typically over 25 different types of rubber available such as natural rubber (NR), styrene-butadiene rubber (SBR), nitrile, ethylene-propylene-diene monomer (EPDM) rubber, fluorocarbon rubber, silicon and many more being used to produce numerous types of products [14]. These rubber generates various sources of waste such as scrap tires, inner tubes, discarded and rejected rubber glove, balloons, rubber bands, shoe soles, mattresses, hoses, seals, gaskets, diaphragms and so on. Table 2 shows the approximate amount of waste rubber being reused, recycled, recovery and landfill or stockpiled globally [14].
Table 1. Applications of waste rubber.

| Application                                                                 | Reference |
|-----------------------------------------------------------------------------|-----------|
| as filler in flooring, shock absorbing mat, roofing materials                | [2]       |
| as an alternative to fossil fuel                                            |           |
| as lightweight fillers                                                      | [3]       |
| as an additives in Portland cement concrete,                                 |           |
| as modifiers in asphalt paving mixtures,                                     |           |
| as crash barriers, bumpers                                                  |           |
| as an artificial reef                                                       |           |
| as energy source                                                            | [11–13]  |
| as an energy source to produce electricity                                   |           |
| being process to recover the oil, steel, vinyl chloride and carbon          |           |
| use in civil application such as for playground and parking lot flooring    |           |
| as filler in road construction                                              |           |
| as modifier for asphalt                                                     |           |
| as a filler in concrete                                                     | [15, 16] |
| as tools for breakwater                                                     | [11]     |
| as floating purposes                                                        |           |
| to produce mats, playground surfaces and trails for athletic used           |           |
| as fuel source                                                              |           |
| as filler in drainage structures                                            | [17]     |
| as filler-extender                                                          | [18]     |
| as an ion exchanger to clear the industrial water from heavy metals.        |           |
| as asphalt modifier                                                         |           |
| as replacement for virgin rubber                                            |           |
| as replacement for carbon black                                             |           |
| as source for mat                                                           | [19]     |
| as workability enhancer                                                     |           |
| as carcass, sidewall and under-tread of tyres                               |           |
| as inner liner for tubeless passenger tyre                                  |           |
| as inner tubes                                                              |           |
| as semi-pneumatic tires                                                     |           |
| as automotive floor mats                                                    |           |
| as adhesive, sealing and tape compounds                                     |           |
| as filler in rubberized asphalt                                             |           |

Table 2. Global estimation of waste rubber management.

| Estimated amount |               |
|------------------|---------------|
| Recycled         | 3 – 15%       |
| Reused           | 5 – 23%       |
| Recovery (energy) | 25 – 60%     |
| Landfill / stockpiled | 20 – 30% |

Tyres are among the major sources of waste rubber due to the large volume produced and problematic to handle due to their durability. Growing number of vehicles worldwide contributes to
approximately 1.4 billion tyres being sold worldwide each year [2]. Each year, it is estimated that, in USA approximately 3.3 million tonnes tyres were generated, whereas in European Union (EU) is 2.5 million tonnes and UK alone is 0.5 million tonnes [14]. Meanwhile, Lo Presti [2] reported that, Europe produced 355 million tyres every year and estimated stockpiles were 5.5 million tonnes. Whereas in 2003, USA generated approximately 290 million scrap tyres. Aslani [20] on the other hand, mentioned that in USA, motorist discarded almost 303 million tyres each years and in 2009 alone approximately 594 thousand tons of scrap tyres were disposed in landfills. Illegal stockpiles of waste tyres were found at the residents’ area which worrying nearby communities of potentially spreading of vulnerable diseases such as Leptospirosis, dengue and etc. Besides landfilling and stockpiles, waste tyres were also being burnt for disposal purpose and causing serious air pollution [8].

Meanwhile, waste rubber gloves also contributed to the amount of waste rubber generated worldwide. After usage, rubber gloves were discarded and dumped as garbage and become a threat to the environment. Rubber gloves was manufactured by fulfilled a strict specifications to ensure it is able to excellently function as a protective barrier to harmful substances. Thus, it has been specially formulated and manufactured with high strength properties and elasticity in order to minimize the possibility that gloves will rupture or tear under stress during used. The basic component in manufacturing glove is latex and additives such as vulcanised agent like sulphur, surfactant; to ensure homogenous and stable compound, accelerator agents to expedite curing process and pigments. All these ingredients has potentially being leached out while the gloves at the landfill and will pollute the soil and water source indirectly.

Furthermore, highly cross-linked thermoset structure of rubber material through vulcanisation process makes rubber glove possessed high resistance properties to acids, alkali, chemical solution and environment agents, which unfortunately it makes rubber glove very difficult to degrade [8]. The increment of waste rubber gloves worldwide relates to an increased demand of glove due to rising and greater concern on health awareness among world population. Societies are concern on safety and hygienic issue that leads to extra cautious on taking self-protection such as wearing glove in food preparation, humanitarian activities, gardening and many more. On top of discarded used glove; higher number of glove disposal also contributed by the high reject rate during the production process due to its strict production specifications and unstable nature of latex [21-22]. Beside tyres and gloves, other rubber waste that contributed to the rubber waste numbers globally is styrene butadiene rubber (SBR) for example from automotive scraps [23], waste rubber condoms [19-24] and industrial rubber waste such as latex foam based materials [25].

2. Development of waste rubber as a filler in a composites material

Many researches paying attention in recycling the waste material especially on polymer based material such as plastic and rubber which resulting a novel sustainable composites. For example, during the 1980s and 1990s significant development and application of utilizing waste tyres were blooming especially in civil engineering [3]. The technologies to recycle waste rubbers are continues to expand as more researchers started to pay attention to this area. Waste rubber were incorporated into various type of matrix such as concrete, latex, thermoplastic and thermoset materials [2, 3, 14, 15, 26–39]. Their investigations has proven that addition of rubber waste has enhanced the properties of the composites such as; improved tensile strength, modulus and elongation at break [19, 21, 23, 39, 40] soft phase of rubber provides flexibility to the composites [22], having better thermal insulation properties [24, 31] and improved impact resistance [41]. Based from this discoveries it shows that use of rubber waste into many potential applications have been proven successful, diversify and the studies are keep emerging. Importantly, the development of rubber based fillers composites benefited for a betterment of the environment. This section will briefly explain the reviewed and development of few researchers in utilizing the waste rubber products into various types of composites such as polymeric based composite and concrete based composite.

2.1 Waste rubber tyres as fillers for composites
The increment number of scrap tyres and stronger environment awareness worldwide makes all parties including federal regulations as well as researchers are giving attention to act for a sustainable approach towards waste tyres abundances. The waste tyres become a serious environmental issue due to their complex structure and component which makes it is difficult to degrade and it requires larger space in landfill and cause landfill limited area for other waste [42]. Many efforts has been conducted to minimizing the waste tyres stockpiling and dumping by treating the waste tyre as a source of sustainable materials.

A comprehensive review was done by Ramarad et al. [11] as they addressed relevant information regarding waste rubber tyres being utilized as a reinforcement material or filler in polymeric material to form a composites. From the review, it found that one of the key element to recycle the waste tyre includes; the rubber needs to be ground and shredded prior being incorporated into the polymer. The ground rubber was then go for surface modification in order to enhance adhesion between the rubber particles with the polymer matrix. Surface modification was conducted either physically or chemically and also by devulcanisation or reclamation process. In this review, they discovered that thermoplastic matrix is commonly being studied by many researchers while natural rubber matrix is also getting extensive attention. On the other hand, little research being conducted for other rubber types of rubber such as butadiene rubber and ethylene propylene monomer (EPDM) rubber. In their review, they highlighted that, waste rubber tyres in rubber matrix is easy to process and its composites acquires an acceptable properties. Review also highlights the lack of studies concentrating on dynamic mechanical, aging, thermal and swelling properties of waste tyre rubber polymeric blends.

On the other hands, Abu Jydil et al. [31] conducted a development of polyester based composites filled with waste rubber tyre crumbs with an intention to be used as insulating material. They conducted up to 40% waste rubber crumb were added into the polyester and being characterized for its insulation properties such as thermal conductivity, water retention, density and thermal stability. They found that, an addition of rubber particles into the polyester matrix has potential to be used as a thermal insulator with the addition of waste rubber tyres has reduced the thermal conductivity and lower the water retention of the composites. The tensile strengths and compressive strength of the composite are higher than currently used insulating materials and it has similar strength to some of the construction materials [32].

Yehia et al. [18] has evaluated the capability of waste rubber to replace certain amount of virgin rubber in natural rubber mix. Their study shows that replacing 10-30% virgin rubber with waste rubber filler does not sacrifice the properties of the rubber mixture. They also found that incorporation of fine waste rubber powder with their surface being chemically treated into the asphalt concrete mixture has greatly improved the performance of the road pavement as the mixture is resistance to crack tip separation.

Reviewed conducted by Shu and Huang [3] reported that the used of waste rubber tyres into asphalt concrete is a successful effort as it improved properties and performance of the mixture. This successful outcomes is a result of good compatibility and interaction between waste rubber particles with the asphalt mixture binder. Lo Presti [2] mentioned in his review that, waste rubber tyres has been used in flooring for playground and sport stadium, in shock-absorbing mat and roofing materials. While, Yang [17] has conducted an investigation of using the scrap tyres as fillers in drainage structures such as for underground and horizontal drain and for ravine crossing. Moustafa and ElGawady [15] highlighted that scrap tyres being used as fillers in concrete to improve the properties such as ductility, damping and impact resistance.

2.2 Waste rubber glove as fillers for composites
The rubber glove industry has grown tremendously over the years to meet the world demands due to rising healthcare awareness globally. This is due to standard of life improved and healthcare reforms. Subsequently, demand for protective products of healthcare safety such as gloves, condoms, latex thread, etc. are also increased [19]. It can be relate with the 26% increment of Malaysia rubber glove
export for the first quarter of 2017 that worth RM4 billion in sales if compared to the first quarter of 2016 which recorded RM3.2 billion [43].

High usage of rubber gloves globally has led to high disposal of used gloves. In addition to the higher number of glove disposal also contributed by the high reject rate during the production process due to strict production specification and unstable nature of latex [19, 40]. Due to their very complex structure and composition that contribute to excellent properties such tremendous elasticity, high resistance to acids, alkali, chemical solutions and environment agents, it makes rubber glove very difficult to degrade consequently creates a serious environmental problem. Glove's ingredients consist of latex (either it is natural or synthetic latex) and vulcanised agents (i.e: sulphur), surfactants, curing accelerator agents and pigments. Thus, dumping into landfill or any open area not only causing piling up but leaching off the ingredients of the glove components will pollute the water and soil.

Rubber glove that possessed many unique properties were beneficial in developing numerous new functional material instead of being treated as invaluable garbage and pollutants but rather a valuable source of sustainable materials. Most importantly, glove are relatively cheap and easy to get thus provide low cost of waste rubber gloves source that lead to low processing cost of the new material [39]. Elastic behavior of rubber glove can be also benefited into numerous field such as damping components, thermal and noise insulator and others. Few literatures has reported that composites with the waste rubber glove fillers shows better mechanical properties such as having better impact strength, better tensile properties and lower stiffness [8, 21, 44-45].

Besides that, there were few other findings that proved, fillers from waste rubber glove has improved the composites properties. For example, blending of filler from waste acrylonitrile butadiene rubber (NBR) glove with epoxidized natural rubber (ENR 50) and NBR rubber glove waste blended ENR 50 has shown improvement of tensile strength, modulus and elongation at break [39, 40]. A study mentioned that, they chose rubber glove specifically NBR waste gloves as a filler in their study because it has excellent puncture and tear resistance, with a reasonable impermeable to many types of chemicals. Thus, these properties will give additional value to the new develop material or composite. In addition using waste NBR glove helps to reduce cost of the material and also easy handling [39].

2.3 Waste rubber condoms as fillers for composites
A study of thermal performance on polymer composite with waste rubber condoms (also known as prophylactics) made from natural rubber as a filler in SBR rubber has been conducted by N.S. Saxena et. al [24]. The author suggested that, result from this study potentially can help in optimizing performance of cryosystem which properties of the composite in low temperature thermal properties can be used to further understand the thermal equilibrium process. The waste condoms were ground into polydispersed particles and later compounded with SBR using two roll mill. The composite was cured in hot press at 150°C and measured for thermal conductivity. The thermal conductivity of the composite was increased with increased of rubber content. A comprehensive review has been conducted by Rajan et al. [19] on recycling natural rubber based waste latex products that include waste condoms. The condoms was first powdered to a size 40 mesh and proceed for reclamation process prior added into raw rubber. This compound reported better in term of processing characteristic.

2.4 Other types of waste rubber as fillers for composites
Zanchet et al. [23] has developed a styrene-butadiene rubber (SBR) composite with utilizing SBR scraps from automotive extruded as its filler. The rubber scraps went for grinding and devulcanisation process through microwave exposure prior being incorporate into the virgin SBR matrix. Revulcanisation of the composite was later conducted by microwave exposure. The addition of SBR scraps giving better tensile strength and elongation if compared to control sample (SBR virgin without SBR scrap). The author also suggested that the increment of tensile strength and elongation due to the devulcanised SBR scraps promotes better interfacial adhesion with the SBR matrix.
Rajan et al. [19] reported that there was a work on using waste latex products as a filler in epoxidized natural rubber (ENR) which helps in reducing stickiness of ENR that give better processability during mixing process. Besides, addition of waste latex product helps reduce mixing time and improved product resistance towards aging. Mathew et. al [44] conducted evaluation on processing aspects, vulcanisation behaviour and dynamic mechanical analysis natural rubber prophylactic (NRP) waste blend with epoxidized natural rubber (ENR) with and without addition of marble powder. The Young’s Modulus and tear strength of ENR increased with increased loading of NRP as well as with the presence of marble powder. But slight decreased at 40 phr of loading NRP. Damping behaviour affected only by the presence of marble powder. The glass transition temperature ENR increase with addition of 40 phr NRP. A study on recycling industrial rubber waste (latex foam waste) has been conducted by incorporating the waste into natural rubber compound for making rubber mat. Addition of the latex waste improved the mechanical properties of this rubberized compound which it has enhanced tensile strength and elongation [25].

3. Conclusion

Use of waste rubber as a fillers into composites, concrete mixtures and other matrices has become one of an attractive field for the researchers to investigate. It is because products that utilizing the waste rubber has high potential to benefit in countless field and at the same time it serve for a betterment of the environment. Recycling of rubber waste into many uses has been proven successful and the study are keep evolving. Researchers also interested in utilizing waste rubber as a reinforcing element in a polymer matrix as well as in rubber matrix. Waste rubber reinforced do provides acceptable mechanical and physical properties to the composites. Beside waste rubber tyres, waste rubber glove and waste rubber condoms, industrial rubber waste are also being recycled. As a conclusion, recycling of waste rubber into a new novel sustainable material has given many positive effect and studies and investigations in this field is rapidly progressing.

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